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Review of the untapped potentials of antimicrobial materials in the construction sector

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ABSTRACT

Microbes can influence the durability of civil engineering structures. Damage caused by microbes (known as biodeterioration) results in compromised structural integrity causing adverse effects on economic and social wellbeing. One key example is that of microbiologically induced concrete corrosion (MICC) due to the bacteria in concrete sewerage pipes which leads to reduced lifespan of sewer pipes. Antimicrobial materials provide a biocidal approach for eradicating the microbes either by inhibiting their growth, or by actively killing them. An ideal antimicrobial material should possess qualities such as sustainability, durability, eco-friendliness, economic viability to avoid the growing issue of antimicrobial resistance (AMR). The literature covering these topics vital to the construction sector is rather scarce. Therefore, this review paper summarises various types of antimicrobial materials currently used in the construction sector detailing their mode of biocidal activity, and their application in structures. This paper also addresses recent developments, demerits and future scope that may aid in employing them expeditiously in the construction sector, particularly to benefit plumbing, and sanitation used in hospitals and high traffic areas and public places including airports, schools and other educational establishments. Overall, the study draws attention to newer antimicrobial mechanisms and provides recommendations for developing new, efficient antimicrobial materials that can provide sustainability and a safe environment to the construction sector.

Abbreviations: AAA, Active antimicrobial agent; AMR, Antimicrobial resistance; AMPs, Antimicrobial peptides; Ag, Silver; BC, Bismuth carbonate; *B. subtilis*, *Bacillus subtilis*; BO, Bismuth oxide; *C. albicans*, *Candida albicans*; CFU, Colony-forming unit; CNT, Carbon nanotube; COM, Complex organic matter; Cu, Copper; DNA, Deoxyribonucleic acid; E. Coli, Escherichia coli; GPC, Geopolymer concrete; GrO, Graphene oxide; H_2O_2, Hydrogen peroxide; HSAB, Hard–soft acid base theory; MD, Molecular dynamics; MICC, Microbiological induced concrete corrosion; MoO_3, Molybdenum trioxide; MTA, Mineral trioxide aggregate; MWCNT, Multi-walled carbon nanotube; NPs, Nanoparticles; LMO, Low molecular organics; PAA, Passive antimicrobial agents; PEG, Polyethylene glycol; PLA, Polylactic acid; QAC, Quaternary ammonium compound; ROS, Reactive oxygen species; *S. aureus*, *Staphylococcus aureus*; SRB, Sulphur reducing bacteria; SOB, Sulphur oxidising bacteria; SWCNT, Single walled carbon nanotube; TiO_2, Titanium oxide; ZnO_2, Zinc oxide; ZO, Zirconium oxide.

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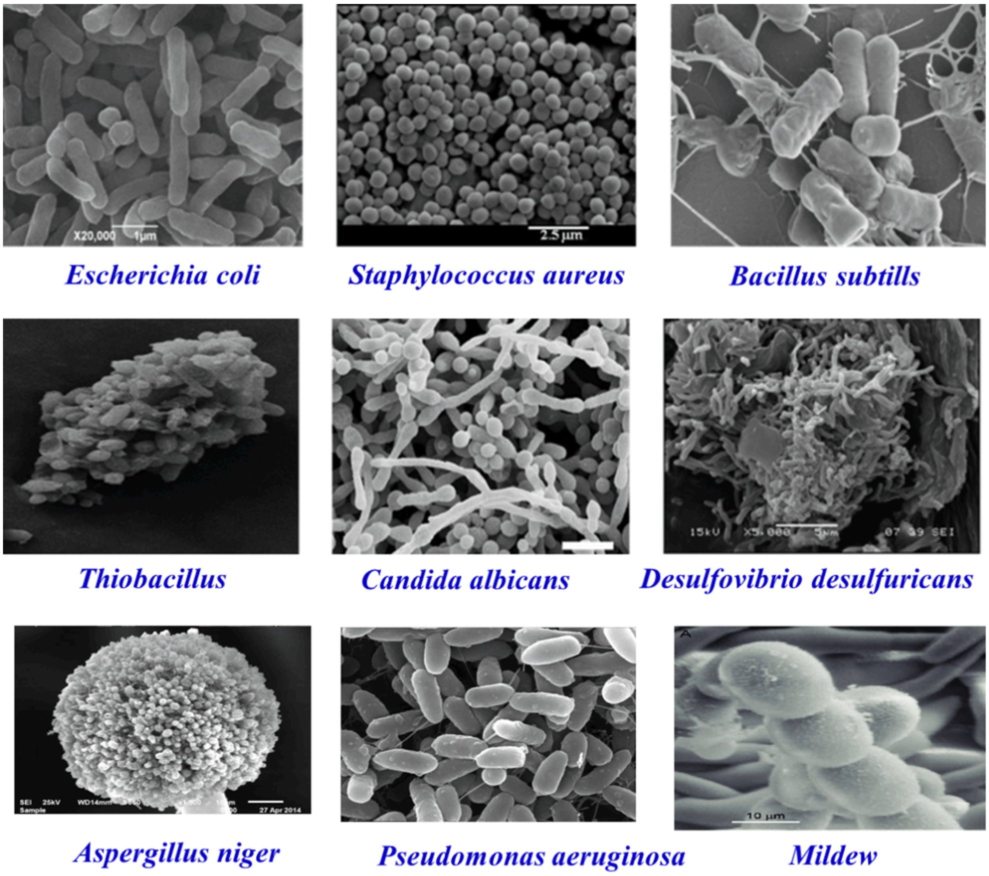


Fig. 1. SEM images of potential microbes prone to affect the quality of human health and to make constructed structures weaker (Prevalence according to the number of times mentioned in the literature) [3–8].

1. Introduction

Microbiological pollution can have a big influence on the deterioration of civil engineering structures. It is important to comprehend the effect and problems associated with this, so that appropriate measures for exterminating them can be adopted [1]. Microorganisms and pathogens such as viruses, bacteria, fungus (mould), and algae tend to grow on surfaces that can be detrimental to the lifespan of structures [2].

E. Coli, *S. aureus*, *Aspergillus niger*, *Bacillus subtills*, *thiobacillus* and other microbes shown in Fig. 1 are some of those which can easily develop in our surroundings and can cause harm to structures and to human health.

Many papers have reported that fungal growth is a major problem in construction industries especially in indoor environments (damp basements, walls, ceilings and window frames) [9]. Also, it has been found that places like toilets in hospitals, airports, train stations, and water storage tanks can host such microorganisms and accelerate their spread [10]. Additionally, many researchers have pinpointed microbiological induced corrosion caused by *Mycobacterium* and *Bacillus* in sewer networks and in the tidal regions due to the fluctuation of wastewater levels and the hydraulic scouring effect [11,12]. Another example for accelerated bio-corrosion and fouling in sewage systems is due to a sulphur oxidizing bacteria (*Thiobacillus spp.*) that generates deadly hydrogen sulphide gas which degrades the lifespan of sewerage pipelines [13]. It has been stated that biocorrosion can drastically reduce the lifetime of sewerage concrete structures down from 100 years to 30–50 years or even less [13]. Historically, to eliminate microorganisms, sterilisation through heat or ionizing radiation have been the most popular methods [14]. However, these strategies last for only a short time since treated surfaces contaminates after some time. Therefore, it is desirable to use antimicrobial solutions or agents that can inhibit, suppress, or kill microorganisms in a more sustained way over a longer period of time.

Antimicrobial agents by definition and design are toxic substances to certain organisms such as bacteria, viruses, and fungi [15]. Over the last few years, there have been tremendous developments in using antimicrobial agents in medical, food, textile and chemical industries. However, the use of these agents in the construction sector is very limited. Triclosan as an antibacterial material was used since 1972, however, it was banned in 2016 by the Food and Drug Administration (FDA) [16]. Graphene oxide (GrO) and carbon nanotubes (CNTs) have been used since 2008, however, their bactericidal effect is still under debatable [17].

Generally, antimicrobial agents used in the construction sector can be administered in three ways (i) by applying a protective layer

Fig. 2. Microbial degradation found in different building materials; (a) fungus growth on wooden ceiling; (b) green growth of cyanobacteria on old stone monuments; (c) corrosion on concrete pipes due to growth of SRB; (d) grey cyanobacteria-dominated biofilms next to red/brown growth of the alga *Trentepohlia*; (e) black mould formed on the ceiling made out on cement plaster; and (f) Orange colouration- Lichen growth producing lichenic acids on bricks [25,28,30,31]. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

like painting (ii) in-situ addition of antimicrobial additives during construction or (iii) addition of antimicrobial agents during the production stage in a factory environment. The first two methods may lead to mishandling of nanoparticles/powders causing contamination of water or soil. They also induce carcinogenic problems. The latter method offers more control in terms of handling of the antimicrobial agents [18]. From sustainability perspective, use of biopolymers can be promising, and some polymers have intrinsic antimicrobial properties, while few others can be modified or doped with other antimicrobial agents like introducing quaternized ammonium or nanoparticles etc. There has been an effort in recent years to introduce nanoparticles (NPs) into construction materials to reduce bacterial colonisation [19]. NPs of ZnO and TiO_2 show effective photocatalytic properties capable to inhibit growth of microbes [20]. Also, the synergistic effort of two or more biocidal materials to eliminate several types of microbes has been lauded as a 'smart' material approach [21] in recent times.

It has been reported that antimicrobial agents achieve their performance by virtue of one or several mechanisms such as electrostatic interactions, production of reactive oxygen species (ROS), or release of metal/metal NP ions [22,23]. Mechanisms like these tend to attack and damage the bacterial cell wall causing cell death of a bacteria. Although many hypotheses, theories and findings exist describing the mode of action of each antimicrobial agent, the exact mechanism by which bacterial killing occurs is still unclear. With the fear of antimicrobial resistance growing, the use of sustainable, eco-friendly, and durable material is becoming more prominent and hence it is important to move forward in selecting the right antimicrobial agent [24].

Civil engineering structures and buildings prone to microbial action/ damage can better be preserved by comprehension of underlying principles of antimicrobial action and the advantages and disadvantages of each antimicrobial strategy. A source providing this coverage cannot be found in the reported literature to date, which was the primary motivation in writing this review. This paper summarises various types of antimicrobial agents used presently in the construction sector, initially highlighting the need for antimicrobial materials in the construction sector while reviewing the detailed problems caused by different microbes in building materials. The paper then explains the nature and mechanism by which antimicrobial materials functions on different substrates. The paper also sheds light on the literature supporting the categorisation of antimicrobial materials based on the source i.e., metals, NPs, polymers, inorganic materials along with the mechanisms and application in the construction sector is discussed. Finally, this paper alludes to the untapped potential of antimicrobials through a critique on certain antimicrobial materials concerning sustainability and highlights the scope of developments in this area. Leveraging the knowledge acquired from disparate fields of biology, materials science and nature, a strategy for effective selection and usage of sustainable antimicrobial agents is proposed by the authors.

2. Need for antimicrobial materials in construction

Microbial activity can influence durability of building materials via a range of biodeterioration mechanisms. The propensity of microbial action depends on the type and extent of microbial growth, type of colonized material and the scale of pollution [25]. For example, in temperate climatic zones, phototrophic eukaryotes, pre-dominantly aerial green algae contribute to the microbial degradation of building materials. However, these phototrophs can colonize most of the building materials in all terrestrial environments, regardless of the geographical area. Likewise, in concrete sewerage pipes, due to the surplus amount of production of sulphuric acid colonisation of sulphur oxidising bacteria can be found which leads to microbiological corrosion [26]. Depending on the type of material and microbes, the symptoms of biodegradation vary. Piotrowska *et al.*, [27] divided the symptoms into two groups: morphological symptoms and changes in the properties of affected material. The changes comprise discoloration and visual presence of organisms, pitting, cavities and disfigurement, friability and fibrillation, structure decomposition, microbiological corrosion, changes in physical (e.g. increased water uptake), electrical (e.g. changes in electrical insulation), optical and chemical characteristics [28,29].

Any microorganism that can hasten the deterioration of building materials requires suitable conditions to develop and grow. For instance, fungi and most bacteria (*E. Coli, S. aureus),* require organic material as a source of energy and carbon, whereas phototrophic organisms, harness sunlight as an energy source and atmospheric carbon dioxide as a carbon source. The degree of degradation depends on roughness, pore size distribution and alkalinity of the materials [28]. Rough and porous surfaces are more vulnerable in facilitating attachment of both airborne propagules and accumulation of nutrient-enriching soiling materials [1]. Therefore, it is important to understand the nature of colonisation and substrate material before deciding on the antimicrobial materials. Some of the specific physio-chemical changes found in different construction materials such as stone, wood, concrete, brick, cement, tiles, glass, steel, etc. (shown in Fig. 2) are discussed in this section. The degradation found in these materials forces the need for usage of antimicrobial material to prevent or kill microbial colonisation on the surface.

2.1. Stones

Most of world's cultural heritage monuments are made of stone [32]. Deterioration of stone instigated by microbes is frequently defined as bio-weathering [33]. Microbial damage of stone depends on intrinsic elements (mineralogical and chemical composition, texture and porous structure) as well as extrinsic elements (water, pollutants, relative humidity, temperature, and biological growth) [34].

Numerous researchers have found phototrophic bacteria such as cyanobacteria, fungi and algae growing on external surfaces of stones [35,36] and *Bacillus Carboniphillus, Streptomyces, Aspergillus,* etc. are mostly found in fissures, cracks or cervices of stone structures [32]. Depending on the type of stone used in the structure, the nature and extent of damage can vary. Some authors have explained that the deterioration induced by green algae activity is mostly assigned to the creation of biofilms, variously-coloured patinas and surface discoloration [25,37]. The first visible changes are alterations in the overall appearance and colour on the surface, with the development of green, brown or pink patches [28]. Additionally, alga cells that contribute to the formation of slime on the surface of stones promote absorption of particles present in the air in the form of dust, soot and spores [38]. Growth of lichen produce acids, called *"lichenic acids",* that damages carbonate stones by causing fissures in them [29].

2.2. Bricks

Bricks are important building materials that have been used as load-bearing structural elements for thousands of years [39]. Bricks can be manufactured manually or mechanically and are highly porous, making them susceptible to microbial contamination [40]. Aesthetic loss, formation of efflorescence, and cracks are some of the major types of damage observed in bricks caused by bacteria, algae, fungi and lichens [39]. Consequently, this increases the need of maintenance and repair work [41]. Many researchers have found the attachment of cyanobacteria cells in small fissures/ cracks on brick surfaces causing structural damage. Algae population inside the buildings developed in the bricks often starts from floors in the washrooms (especially near the windows), whereas lichens grow primarily on the roof tiles made of clay [27]. It has been found that *Cladosporium, Aspergillus, Penicillium, Alternaria, Stachybotrys and Helminthosporium* are the most common outdoor moulds, while *Cladosporium, Alternaria, Aspergillus and Penicillium* are common indoor moulds that can easily grow on the porous bricks with an appropriate moisture and humidity conditions [42].

2.3. Wood

Wood and wood products are commonly used in floors, plates, walls, ceilings, interior fittings and furniture [43]. Due to its biodegradable property, it is highly susceptible to microorganisms, fungi, algae, insects, etc. Wood damage by bacteria and fungi involves degradation of cellulose, hemicellulose and lignin. This leads to aesthetic deterioration of the surface (peeling, delamination, discoloration) and, above all, structural and mechanical changes (reduced strength, hardness) [44,45]. *Penicillium, Aureobasidium, Aspergillus, Acremonium* and *Sphaeropsidales* are some of the major fungi found on wooden surfaces [42]. Brown rot fungi (*Poria vaporaria, Serpula lacry- mans*) considerably decrease wood's strength to 30 % of the initial strength within 6 months, which is particularly dangerous in the case of structural elements. In turn, *Corticiaceae* fungi causes superficial wood degradation, and thus they pose smaller risk to the strength of structural elements [27,44]. Depending upon the locations and seasons, growth of fungus found on wooden surfaces vary. Piotrowska *et al.*, [27] observed that in Germany, the total count of the microbial growth on wooden surfaces were mostly during autumn season where humidity is relatively higher than other seasons.

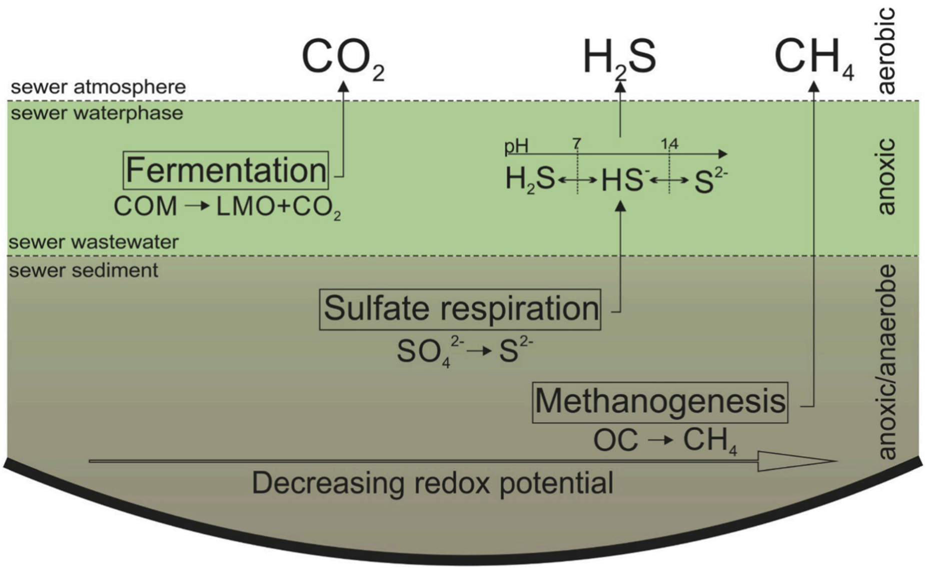


Fig. 3. Schematic diagram illustrating MIC in sewer concrete pipes [57].

2.4. Portland cement and mortar

Cement is the second most used material in construction. Literature reports that calcite and silica from the cementitious substrate plays an important role in bio receptivity since it can favour colonization of microorganisms [46,47]. The consequences are mainly aesthetic: green, black or red stains develop, sometimes very quickly, on infrastructures, depending on a variety of climate, environmental and architectural parameters [48]. Fungi can also participate in this kind of attack. In this case, beside aesthetic alterations, building materials can suffer mineralogical and microstructural damage such as reducing pH, formation of cracks and corrosion [26]. Irrespective of indoor or outdoor conditions, microbial growth can be observed on the mortar especially when utilised as connecting blocks between stones or bricks [28]. These deterioration can be easily pictured from all the heritage monuments that are built with plaster and lime mortars [49]. Both algae and cyanobacteria easily grow on walls made out from cement plasters and can be seen as greyed colour patches in dry region whereas humid areas are of green colour [37]. Growth of cyanobacteria on the mortar surfaces converts calcium carbonate into gypsum and this traps the particulates from the atmosphere which lead to blackening of the surfaces called as "black crusts" and it is found majorly in most polluted areas [1].

2.5. Concrete

Concrete undergoes microbiological degradation that can cause serious structural damage. One of the well-known examples is biodeterioration caused by the microbes leading to biocorrosion or microbiological induced concrete corrosion (MICC). It has been observed that the loss of structural capacity with time is caused mainly by chloride ingress through six transport processes: adsorption, diffusion, binding, penetration, capillary action and dispersion which leads to steel corrosion (loss of effective cross-section of steel), concrete cracking, loss of bond (aggregate-hydrated cement paste) and spalling [50,51]. Among these causes of structural degradation, it has been noted that deterioration arising from biological sources is significant in harsh environments which is referred to as microbiological induced concrete corrosion (MICC) [52]. Various microorganisms have been implicated in MICC; these are commonly categorised in groups according to their metabolic capabilities. The main groups related with MICC include sulphate-reducing bacteria and archaea, thiosulphate-reducing bacteria, acid-producing bacteria, iron-oxidising bacteria, iron-reducing bacteria, nitrate-reducing bacteria, and methanogenic archaea [53,54]. Therefore, MICC is a complex phenomenon that can be triggered by several microorganisms with different metabolic capabilities. MICC is a result of sequential activities including biogenic controlled sulphate reduction and redox reactions [26,55]. It is instigated by sulphate reducing bacteria (SRB) (e.g., *Desulfovibrio*) in anaerobic conditions whereby biological reduction of sulphate ($SO4^{2-}$) and organic sulphide (S^2) to hydrogen sulphide (H_2S) are produced at the bottom part of the pipes [10]. Afterwards, the oxidation of hydrogen sulphide into sulphuric acid, and degrades the concrete [56]. Internal cracks and pitting corrosion of concrete caused by the formation of calcium-containing products not only increase the surface area causing microbial degradation, but also reduce structural integrity and thus shorten the life of concrete structures [13]. Grengg *et al.*, [57] explained that microbial degradation caused in sewer pipes occurs in two phases (Fig. 3); (i) Initial fermentation process causes the transformation of complex organic molecules (COM) into low molecule organics (LMO) accompanied by CO_2 production; and (ii) LMO are then consumed during the sulphate respiration of sulphate-reducing bacteria, as well as during the methanogenesis of methane-producing bacteria under strongly anaerobic conditions, resulting in the production of sulphide species (H_2S, HS^- and S^{2-}) and methane (CH_4) and CO_2. Researchers have found that addition of zeolites comprising silver and copper ions suppressed *Thiobacillus sp.*,

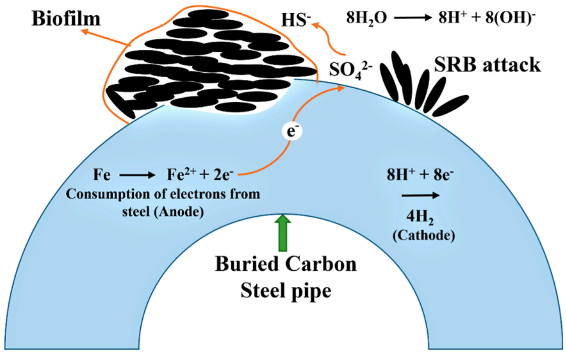


Fig. 4. Microbiological induced corrosion on steel due to SRB attack [65].

E. Coli and *Salmonella* at an optimal dosage of 1 wt% [19]. Some other researchers have noted that adding calcium formate to concrete prevents the growth of sulphur-oxidizing bacteria (SOB) and prevents biological corrosion in marine systems. Numerous studies have revealed that the use of nickel and tungsten protects concrete against *Thiobacillus thiooxidants* [13]. A few researchers have also reported that the use of anti-bacteriostatic material can limit the microbial growth on concrete surfaces in aggressive sewer environments effectively [57]. Also, Hashimoto *et al.*, [58] and Grengg *et al.*, [59] suggested geopolymer concrete as a green sustainable approach towards MIC in sewer pipes due to their high resistance to the acidic environment.

Gaylarde *et al.*, [1] also indicated that the presence of calcium in concrete instigates the growth of fungi that can significantly reduce pH. Fungi increase this physical damage in the concrete structures by etching and extending hyphae that penetrate inside surfaces, thus resulting in enlargement of the already damaged area and an increased porosity [28]. The growth of fungus (*Fusarium*) in humid areas can be identified by black biofilms on their surfaces [60]. It has been reported that concrete with a high water/concrete (w/c) ratio is more vulnerable to fouling by algae. In addition, larger surface roughness and capillary porosity have been observed to increase primary bio receptivity of concrete to filamentous algae as well as fungi species [61].

2.6. Structural steel

Rebars, window walls, roofing, interior decorations, railing, staircase, pipelines etc. are some of the major applications where steel is used in construction [62]. The most used steel in construction are mild steel and stainless steels. Different stainless steels have different alloy elements and surface properties which could have significant influence on bacterial attachment to the surface and thus might result in different microbiological corrosion behaviour [63]. Commercially available steels such as mild steel and stainless steel can corrode in moist environment and this can lead to the growth of sulphur reducing bacteria (SRB) [64]. Localised corrosion like pitting, crevice corrosion, inter-granular corrosion, fatigue etc. are some of the problems associated with microbiological induced corrosion (MIC) [1]. It is inferred that once SRB interacts with water and metal; it creates a layer of molecular hydrogen on the metal surface. The SRB then oxidizes the hydrogen while creating hydrogen sulphide, which contributes to corrosion which has been illustrated in Fig. 4 [65]. Bacteria can release aggressive metabolites, such as organic (acetic, succinic, isobuteric, etc.) or inorganic acid (sulphuric) that causes corrosion and degradation of steel [66].

Gallionella, and filamentous bacteria in the genera *Sphaerotilus, Crenothrix, Leptothrix, Clonothrix, and Lieskeella* [84] are some of the known iron and manganese oxidizing bacteria that oxidise ferrous to ferric iron ($Fe^{2+} = Fe^{3+} + e$), catalysing the deposition of tubercles, especially on stainless steel weld seams [66] thus leading to excess chloride ion concentration causing corrosion. Production of sulphuric acid, hydrogen embrittlement, cathodic depolarisation, reduction of ferric to soluble ferrous iron, slime and acid production, formation of oxygen concentration cells, etc. are some of the corrosion mechanisms that are caused by different types of bacteria on mild steel when exposed [67].

2.7. Paper

Paper has been used as a building material for many years particularly as cardboard, wallpaper, paper-concrete, paper nanocomposites, cardboard tubes, etc. [68]. They are also used as wall panels, thermal insulation panels, hollow core panels, architectural and decorative blocks [69]. Several researchers have found that paper products are more susceptible to airborne fungus like *Aspergillus* and *Penicillium* [70]. A few researchers reported that *Cladosporium, Stachybotrys E. Coli, Salmonella cholerasuis, Bacillus cereus, S. aureus*, and *C. albicans* growth is often observed on paper products [71]. These microorganisms exhibit cellulolytic, proteolytic and amylolytic activity to produce acids, which help in the degradation of paper material [42].

2.8. Glass

More than 150 years research on microbial interactions with glass has been carried out with the aim to understand the influence of microbial growth. Usually, biofilm is formed on the surface of glass that may cause alteration in the form of biophysical and

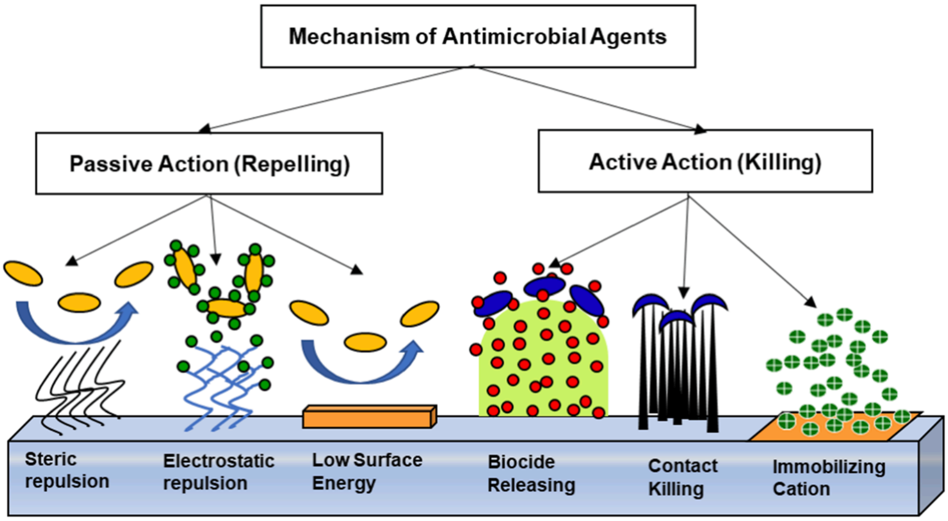


Fig. 5. Mode of action of antimicrobial materials.

biochemical processes [72]. Biophysical alterations are due to mechanical stresses developed during the growth of the microorganisms on the surfaces which lead to fissures or cracks on the glass surfaces. Biochemical alteration is defined as a process where chemicals produced by microorganisms cause leaching of elements from a glass surface [73]. Green biofilms caused by growth of cyanobacteria can successfully colonize and develop in glass materials [27]. At ambient temperature and pressure, glass has a natural tendency to transform into the mineral phase, and undergoes weathering and other decay processes such as the microbial colonisation [74]. Colour change of the surface due to fungal or cyanobacterial growth and biogenic minerals deposition because of the microbial metabolism on the glass surface are examples of deterioration categories observed on glass surfaces [74]. Further they lead to in situ crack formation, pitting, and chipping of the glass in the presence of various micro-organisms [75].

3. Mechanism of antimicrobial action

Antimicrobial agents can be classified as passive or active (Fig. 5). Passive agents are the one that prevents the growth of microbes whereas active agents can eradicate microbes completely [76]. Antimicrobials can be both natural and artificial [77,78].

3.1. Passive antimicrobial materials

A passive antimicrobial agent (PAA) diminishes protein adsorption. In doing so it precludes the growth of microbes on the surface. PAA can only repel microorganisms and not eradicate microbes wholly [22]. The mechanism with which PAA works (see Fig. 5) can be further classified as: (a) Steric repulsion (b) Electrostatic repulsion and (c) Repulsion due to low surface energy. Typically, PAA would be non-adhesive coating materials and are considered as antifouling agents. Therefore, hydrophobic, and negatively charged microbes can be repelled easily by a hydrophilic, negatively charges and low surface energy material [79]. It is reported that PAA fails in its functionality due to oxidative degradation over time and loses its durability [80].

3.1.1. Steric repulsion

Steric repulsion arises from the repulsive forces possessed by the antimicrobial polymer chains [81]. Zwitterionic polymers such as Poly(carboxybetaine) (PCB) and poly(sulfobetaine) (PSB), etc. are some of the most prominent examples of antifouling agents which can be classified in this category. They do so by forming a hydration shell due to the steric hindrance effect [82], Sumdani *et al.*, [83] examined the effect of multi-walled carbon nanotubes by dispersing it in epoxy composite and observed that thermal properties and antifouling properties of the composite increases extensively due to steric repulsion of homogenous dispersed carbon nanotubes in composites. Also, steric repulsion is interpreted as an impact aroused by an adverse variation in free energy related to dehydration and confinement of the flexible polymer chain [23]. A few other researchers have pointed out that polymers like PEG shows antifouling properties through steric repulsion accompanied with surface hydration [84].

3.1.2. Electrostatic repulsion

Electrostatic interactions arise from attractive or repulsive forces between charged particles [84]. The electrostatic interactions between the bacteria and an antimicrobial substrate surface are usually repulsive since surfaces normally tend to be negative due to conditioning by reactions with oxides from the surroundings. Bacteria is negatively charged due to the cell constituents containing carboxyl, phosphate and acidic groups [85]. Electrostatic interaction between the substrate and microbes involves various factors such as pH, type of substrate, ionic strength and the electronegativity of the combination etc. [85]. Qiu *et al.,* [19] noticed that the

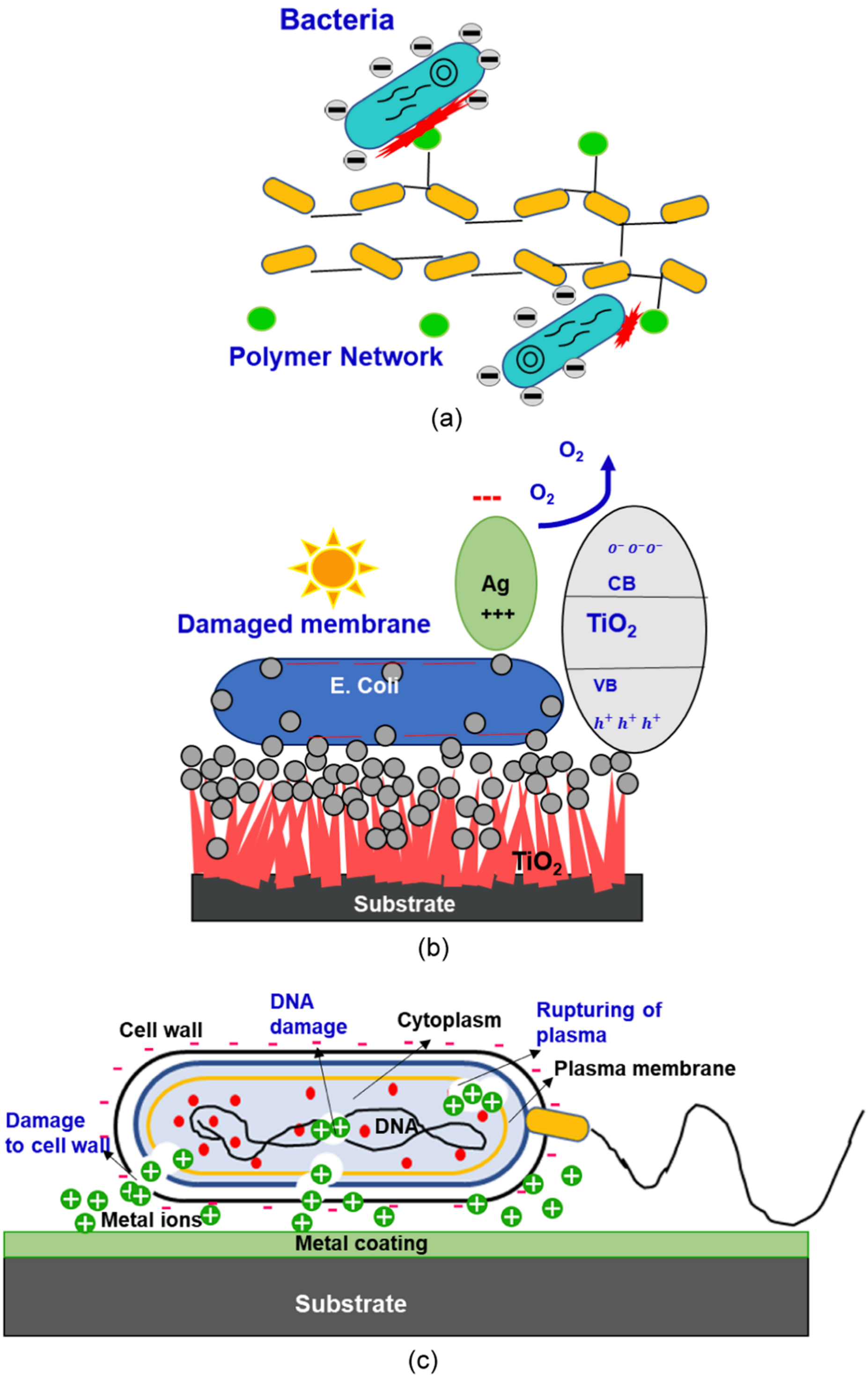


Fig. 6. Various types of biocides; (a) Polymer sterilisation; (b) Photocatalyst sterilisation; and (c) Metal sterilisation.

electrostatic repulsion between the –COO– units of the substrate and bacteria was more when pH of substrate is more than pKa. However, some investigations have also reported that antimicrobial hydrogels having pH of 7.4 pH turn out to be more hydrophobic and more resilient to *Staphylococcus epidermidis* (*S. epidermidis*) [86,87]. Many researchers have speculated that antimicrobial peptides (AMPs) with low molecular weight which have protonated primary amine groups tend to confer the desired bioactivity profiles in supressing the growth of microbes [88,89]. Metal nanomaterials such as copper, zinc, silver, zirconia, etc. which are positively charged can attach to the negatively charged bacterial cell via electrostatic repulsion and can disrupt the DNA of the bacteria [76]. Therefore, addition of these antimicrobial polymers, metals or metal nanomaterials as a paint can prevent bacterial adhesion on the substrate and provide antifouling properties.

3.1.3. Low surface energy

Controlling the surface energy provides a good mechanism to prevent the microbial growth forming a colony [90]. Researchers have speculated that antimicrobial agents like polymers must possess a flexible linear bone chain with low intermolecular interaction forces between the constituents [90]. Many researchers have noticed the similarity of working principles in hydrophobic and superhydrophobic polymers, hydrogels having low surface energy with *Lotus leaf effect* wherein bacteria do not adhere on the substrate, but roll off easily [91,92]. A plethora of research has identified that nanoscale roughness materials demonstrates low surface energy which makes them superhydrophobic [93–95]. A clear correlation between hydrophobicity and antibacterial performance has never been established to date [96].

3.2. Active antimicrobial agents (AAA)

Active antimicrobial agents (AAA) can eliminate microbes instead of just preventing their growth. They can be categorised as (a) biocide releasing agents; (b) contact killing agents and (c) immobilizing ion agents. Positively charged quaternary ammonium is the most extensively utilised AAA, that can react with the bacteria cell wall leading to the release of cytoplasm through the cell membrane, causing death of bacteria [22,97]. Metals and nano particles such as carbon nanotubes, iron (III) oxide, zinc oxide, magnesium oxide, silver, gold, copper and copper oxide, calcium oxide, titanium dioxide and cadmium oxide are specifically considered as AAA [76,98,99].

3.2.1. Biocide releasing

A popular method to achieve a biocide releasing mechanism is by impregnation of materials [91]. Impregnated materials can work as carriers of biocides that can be discharged near to the cell in high local concentration causing instant death of microbes [100]. Biocide releasing metal ions (silver ions), polymer ions (triclosan), chlorine, nitric oxide, photocatalytic substances etc. are some of the major examples that have been used during the past few decades as shown in Fig. 6 [101]. Researchers have elucidated that applications of coatings using one or more biocides as a monomer in a polymer were discovered to lessen the growth of *E-Coli, S. aureus* and *Bacillus subtilis* [91]. One of the challenging issues that many researchers have noticed in using biocide releasing polymer-based coatings as that it often changes their surface properties after releasing biocides [102]. Therefore, they are generally combined with other polymeric substances that can frequently renew the surface after washing away all the attached microbes. Such coatings are generally used in ship building to protect hulls as well as marine buildings where biofouling can be quite severe [101]. Although release of biocides kill microbes instantly, it is seen as a toxic practice [80]. Additionally, a synthetic biocide may become sedentary over time which can stimulate antimicrobial resistance (AMR). Owing to its toxicity, the EPA of the USA has restricted and categorised all the biocide releasing materials based on its type and application. Nowadays use of natural biocides including extract from betel leaves, green tea, turmeric, garlic etc. are becoming popular. It is also seen to have successfully suppressed planktonic growth [103].

3.2.2. Contact killing

Contact killing is achieved through a mechano-biocide action wherein biocidal activity occurs by stress exerted by the sharp spikes protruding out of a surface which causes fracture of the cell wall [104]. It has been suggested that contact killing occurs due to (i) spacer effect-where a bacterium becomes stuck between two spikes; (ii) polymers having positively charged QACs, can detach phospholipids from the cell membrane and thereby kill the bacteria [80]. In simple words, the adhered bacteria will be destroyed by serious membrane interruption via a polymeric spacer effect, ion-exchange mechanism and phospholipid sponge effect [81]. The presence of a charge density level on positively charged biocidal surfaces has offered additional insights into the interaction of such surfaces with bacteria. The ion exchange mechanism is known to be more prevalent for contact killing of bacteria whereby the exchange of divalent cations present on outer membrane of bacteria can occur with cations present on the charged surface of the antimicrobial agents [105]. This proceeds to damage and create loss of natural counter-ions of bacteria, causing immediate cell death. Many researchers have found that the mechanism of killing bacteria by contact killing is valid for both gram positive and gram negatively bacteria, however, not many studies exist for killing yeast and viruses [106]. Polymers having covalently bonded antimicrobial moieties, such as immobilized QAC, guanidine group, possess the unique feature of bacterial 'contact-killing' by destroying the cell membrane of the negative charged bacteria [104,107]. *N*-chloramine based contact killing antibacterial agents have achieved substantial research attention over the past years. Kaur and Liu [81] reported that *N*-chloramine initially attacks through chlorination of proteins of bacteria and penetrates inside bacteria to oxidise the vital part of cells resulting in cellular death. Similarly, many researchers have reported that release of metal ions such as copper or silver may also cause immediate cellular death when bacteria comes in contact with a surface [108,109]. Mathew *et al.,* [110] evaluated the contact killing mechanism of copper using laser interference lithography and observed that the influx of copper ions into the cytoplasm of the bacteria is the key factor of killing bacteria on a dry surface. Additionally, this contact killing mechanism is slow when the surface is wet. Few other researchers have also reported that plasmid DNA is completely degraded after cellular death preventing the transfer of resistance between organisms [111]. Recent studies have reported that the use of physical contact killing mechanism on nanostructured surfaces such as nature inspired modified surface (cicada wings, dragon wings, lotus leaf, etc.) is a promising strategy for curtailing the spread of bacteria [112,113].

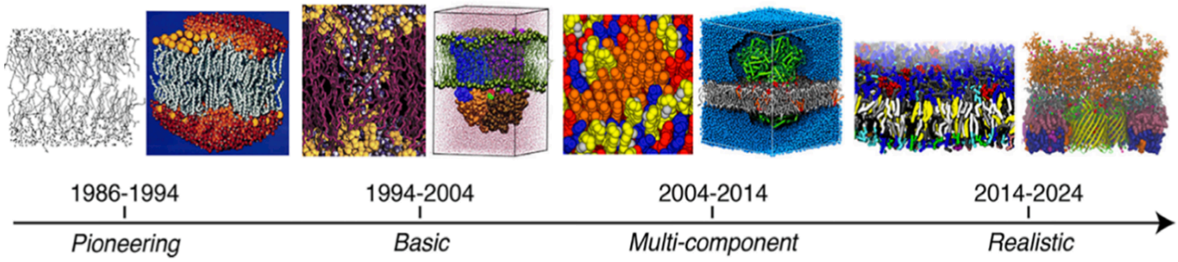


Fig. 7. Evolution of molecular simulations over past 30 years [125].

3.2.3. Immobilizing cation

Immobilized ions on the substrate become activated while encountering bacteria. Researchers have identified that unlike other antimicrobial agents, use of immobilized ions potentially increases efficacy of antimicrobial action [114]. These agents disrupt membranes of microbes to kill them [115]. There are three major strategies for immobilisation of antimicrobial agents to the surface [114]: (i) physical adsorption which is governed by physisorption of antimicrobial agents with strong non-covalently but interaction with the substrate, (ii) "graft to" which occurs through the covalent bonding between the substrate and antimicrobial agents and (iii) "as formed" development of substrate which contains antimicrobial agents at the time of production, i.e., it is unlike other substrates which require an activation process to generate amine, carboxylic acid that can kill bacteria [14]. Over the past few years, a variety of immobilising agents have been discovered which includes silane, amine, peptides, quaternary ammonium polymers, chitosan and silver nanoparticles that are found to be effective in killing bacteria [116–118]. It is reported that one of the key potential drawbacks of this method relates to the fact that the surface becomes fouled by the same bacteria that it kills, and this causes serious maintenance problems [114]. However, Spagnul *et al.,* [119] used a very promising alternative to conventional immobilizing agents. They used a Photodynamic antimicrobial agent which relies on employing a photosensitive agent. This becomes activated by a non-thermal visible light of appropriate wavelengths to generate high localised concentrations of reactive oxygen species (ROS). This in turn deactivated microbes. Nowadays, there is another approach becoming popular especially in the medical field which is about immobilizing bacteriophages on surfaces [80,120]. Bacteriophages are viruses (immobilised on the substrate) that infects and kills bacteria without any negative effect on the substrate. These are more efficient and relatively cost-effective than other antimicrobial methods [121]. However, this method has yet to enter the field of construction and is a worthy direction to pursue.

3.3. Simulation studies on antimicrobial materials

An atomic scale understanding of antimicrobial performance has continued to remain a very fertile area. The simulation-based studies can be classified into analytical and computational (numerical) models [95]. Analytical models involve equations based on surface energy considerations, thermodynamic equilibrium and energy minimisation are formulated and solved by introducing geometric constraints relating to both bacteria and nanostructured surfaces [122]. Computational models on the other hand, involve bacteria – substrate interaction using numerical solvers such as the finite-element method or molecular dynamics [123]. Having said that, molecular dynamic (MD) simulations can deliver molecular level specifics and information that are essential to understand the bactericidal activity [124]. It has been proposed that the interaction between positively charged antimicrobials and negatively charged bacterial cells is the main cause for bactericidal activity [88]. In principle, molecular dynamics (MD) can describe the interactions between all components at atomic resolution, acting like a "computational microscope" [125]. Use of MD simulations for understanding the interaction in important parts of a cell started from the 1990s [125]. Thanks to developments in science and technology, we are now exploring a change from simulations of simplified, model membranes toward multicomponent realistic membranes with the efficient use of graphical processing units (GPUs), together with the development of accurate atomistic and coarse-grain (CG) models [126,127]. Fig. 7 illustrates the transition in complexity of molecular modelling from the 1990 s to present times.

Some of the commonly used MD softwares are LAMMPS, NAMD, AMBER, GROMACS, Abalone, Ascplah designers and Materials studio [125,128]. The quality of simulation depends on the quality of force field (FF) or the so-called potential energy function or simply called material constitutive model, i.e., set of parameters indicating how particle/molecules interact with each other. Many researchers have reported that a possible way to extend molecular modelling and bridge it with experimental methods is to use a coarse-grain model (CG) [129]. Recent advances in computation and molecular FFs have allowed for more meaningful modelling of realistic simulations wherein cell modelling at mesoscale has been performed to understand the cellular mechanics [130]. To develop an MD simulation, assumptions must be made as to what forces govern the phenomenon being modelled. Gravity, adhesion and interfacial energy gradient are some of the important parameters which aids understanding of antimicrobial structured surfaces exhibiting different frictional behaviours and correlating it with their bactericidal performance especially in the case of mechano-bactericidal surfaces (nano-pillars) [95].

Additionally, specific parameters such as hydrophobicity, low surface energy, high surface roughness, etc. are necessary to make a surface to behave antimicrobial [131]. However, these basic parameters alone are not sufficient to robustly design antimicrobial materials with great therapeutic values. There is no simple, linear relation among antimicrobial materials, intrinsic factors and their bactericidal activity [132]. As a result, innate design rules for novel antimicrobial materials are hard to deduce. The wide variety of

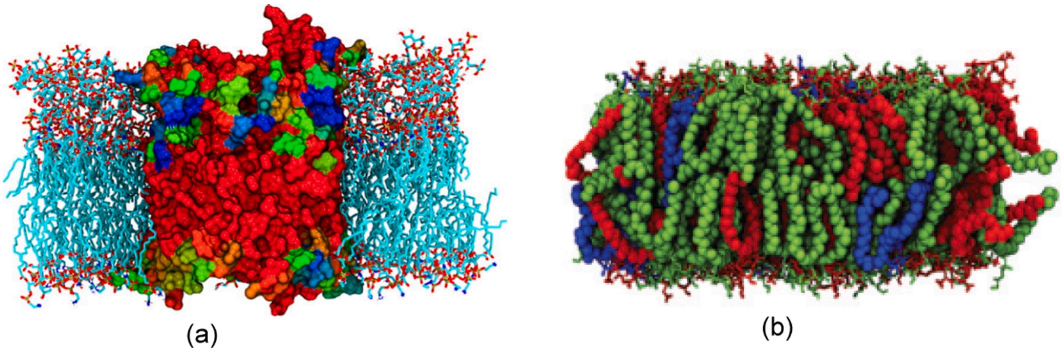


Fig. 8. Illustration of outer membrane layer of (a) *E. coli* and (b) *S. aureus* [135,136].

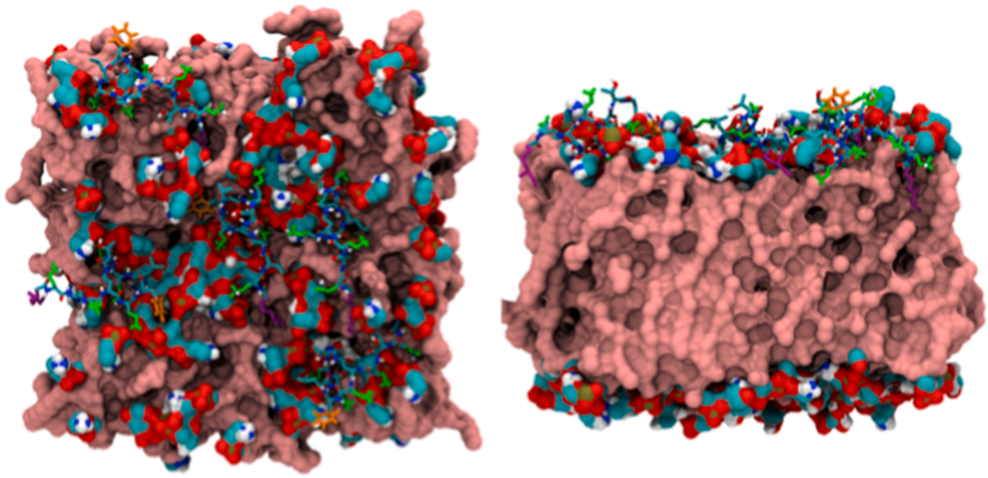


Fig. 9. Top and side view of the AMP interaction with outer membrane of *E. Coli* [135].

sequences, structures and mechanisms implies that there are always numerous aspects involved in the killing of bacteria [9]. In this section, we briefly review some of the investigations conducted by various researchers using MD simulations to study the bactericidal action of antimicrobials.

The exploration of such detailed and realistic simulations has been carried out primarily in the medical field whereby interactions of different antibiotics with subsequent bacteria/virus/fungi are carried out [133]. Fig. 8 shows basic and detailed MD simulation models of the outer membrane layer of *E. coli,* and *S. aureus* having phospholipids and proteins. This type of detailed modelling for each bacterium is important to predict possible ways in which a bacterial death occurs while coming in contact with an antimicrobial material.

Berglund *et al.*, [134] reported the MD study of antimicrobial peptide (polymyxin B1) interaction with both inner and outer membranes of *E. Coli* bacteria. It was revealed that peptides insert readily into the inner membrane (driven by electrostatic interaction), whereas the interaction with outer membrane is more complex and no physical damage was observed to cause a bactericidal effect [135]. Fig. 9 shows the top and side view of AMP interaction with the outer membrane layer of E. Coli.

Recently, the use of MD has become more prevalent to study the mechanics of nanopatterned surfaces [137]. The bactericidal activity observed from the experimental techniques such as the scanning electron microscope, atomic force microscope and time-lapse fluorescence imaging, are investigated in greater details using MD simulation to investigate their temporal evolution and spatial distribution [138,139]. It was reported that combined experimental and simulation investigations can analyse a substrate in an hour instead of requiring overnight incubation as in CFU counting [138]. Simulation and experimental results both have shown strong correlations between surface morphology/wetting and cell damage of bacteria [132,140]. Most of the studies used *in-silico* approaches to simulate the bactericidal effects of nano-patterns and to identify the favourable range of geometrical aspects with different bacteria for designing artificial antibacterial surfaces [141–143].

Velic *et al.,* [144] analysed the mechanics of adhesion-driven envelope deformation by computational methods, namely 3D finite element analysis using Abaqus, to study the magnitude and location of critical strains induced in cicada wings. It was concluded that the numerical models reaffirmed adhesion-driven deformation and death, as originally proposed by *Pogodin* (biophysical model-passive adhesion force and van der Waal's force) and cell wall deformation is increased by adhesion to nanopatterns with smaller pillar radii and spacing. Some studies have also elucidated that sharper peaks would improve the antimicrobial effect of the nano-

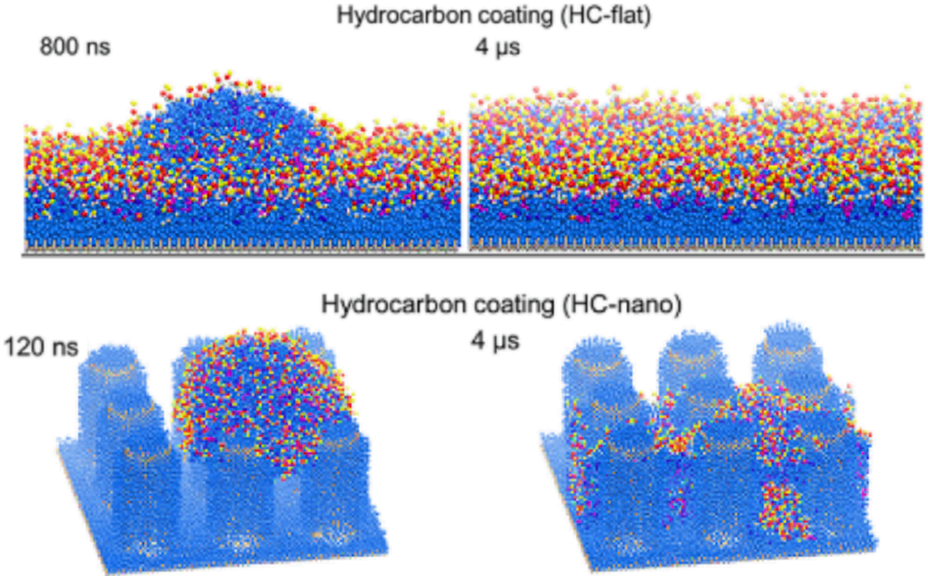


Fig. 10. Interaction of bacteria on flat and nanopatterned surface [138].

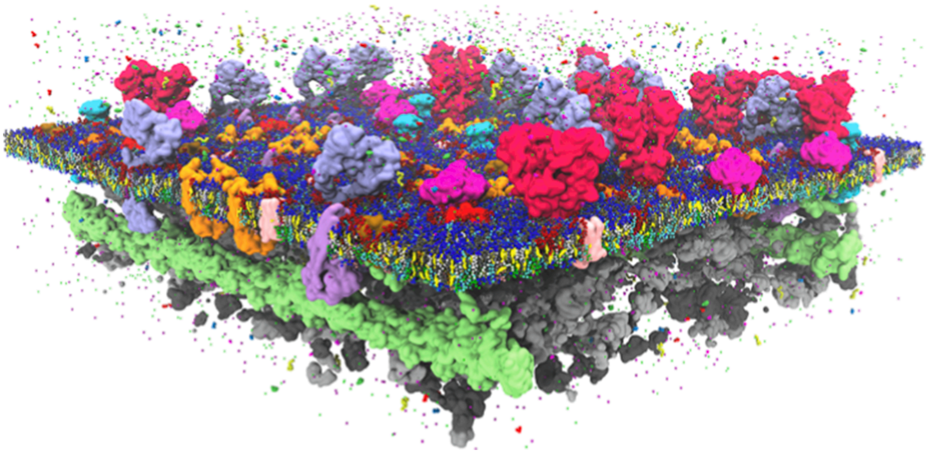


Fig. 11. Real time full cell MD simulation of a cell [125].

patterns [140,145]. The aspect ratio was identified to be a more vital constraint to enhance the bactericidal effects against gram-negative bacteria while spacing was found to be more influential against gram-positive bacteria [141].

Non-linear FEM analyses (Abaqus) were explored to simulate the interaction of different bacteria species with NPs to identify the role of geometrical features in inducing optimum bactericidal effects [141,144]. It was observed that the equivalent von-Mises strain in the outer wall of the bacteria was found to surpass the experimentally assessed threshold rupture strain. This indicated cell rupture which was linked to the extent of deformation of the cell wall while coming in contact with the NPs [141]. Sibilo *et al.*, [138] performed MD simulations following the 4-to-1 coarse-grained (CG) procedure described in the Martini v2.0 force field (Fig. 10). They studied the adhesion/destruction by assuming the bacterium as a small lipid vesicle. They introduced Coulombic and van der Waals forces to minimize intra-vesicle and surface interactions and to avoid collapse of the molecules. It was inferred that surfaces coated with hydrocarbons showed a superhydrophobic Cassie – Baxter wetting state causing complete rupture of bacteria but relatively less rupture when its surface was coated with fluorocarbon [146].

Additionally, from the simulations it was noted that the cumulative intermolecular force associated with van der Waals, hydrophobic, and electrostatic effects between the vesicle and the surface was sufficient for complete disruption of the phospholipid of bacteria [147]. Literature also reported the effectiveness of MD in revealing the chemistry of super hydrophilic surfaces like zwitterion polymers, polymethyl methacrylate (PMMA) possessing antifouling capability and electrostatic interaction with fungus [148,149]. It was also noted that a strong repulsive force exists between the substrate and fungus.

The ability to individually manipulate and simulate a wide range of physical parameters via a simulation-based approach paves the way to a robust, unified theory of the antimicrobial materials. Eventually, MD simulations aids in the reduction of tedious experimental procedures and saves time. By mimicking the actual phenomena of bactericidal activity, it can help in a detailed understanding of the nature, geometry, biological, and environmental parameters that are responsible in governing the antimicrobial action. Considering the progress not only in complexity but also in system sizes that can be simulated with particle-based models, it is not easy to predict that a full cell simulation at near- atomic resolution (Fig. 11) is feasible within the next few years that can help in understanding and predicting the actual scenario between microbes and antibacterial surfaces. Such types of studies are yet to be explored in the field of construction. Additionally, it will also predict the altered properties of subsequent building materials and their useful life before biodeterioration.

4. Current practices to use antimicrobial materials in construction sector

Antimicrobial performance in construction is currently achieved by one of three ways (i) by providing paint or coating on the finished surface after the basic construction; (ii) by mixing of inorganic additives (such as metallic nanopowders e.g., copper) into concrete/mortar (site production) during construction (iii) by mixing antimicrobial agents during fabrication of constituent materials such as cement, bricks, tiles, blocks during their factory production. The first method is more popular, although, providing a protective layer like paint or coating on tiles, walls, doors, pipes, etc. not only incurs initial expense but also additional recurring costs of at least $10 to $20 per square metre [23]. It can also be argued that the use of metallic powders as additives in a concrete mix at a construction site can lead to issues such as mishandling of metallic nanopowders leading to accidental inhalation causing carcinogenic problems or their accidental release into soil or water can cause critical environmental issues including groundwater and agricultural contamination. A factory environment would theoretically provide better control to avoid such accidental handling issues and would guarantee that the mixing is done in an optimal manner, although, research in this area is still scarce.

A common way to prevent biological contamination on any material is to clean it frequently by using disinfectants, cleaning agents etc., however, use of such chemicals can cause environmental issues including the AMR [91]. In a building, a wide range of products such as doorknobs, tiles, walls, door, taps, water tanks, sinks, urinals, wash basins etc. are some of the important areas where antimicrobial coatings are cautiously applied. The idea behind applying antimicrobial coatings is to eliminate the possibility of letting bacteria rest on the surface and thus to avoid the formation of biofilm [21]. There are basically two main categories of antimicrobial coating that are used (i) bacterial resistant coating which inhibits initial attachment at early stages; (ii) bactericidal which can kill the bacteria landing on it. Further these are classified and chosen depending on the mode of action, type of prominent microbes, location, and condition of materials. Nowadays, with emerging and advancing technologies, “smart” coatings are gaining importance [150]. Smart coatings can adapt to tailor antibacterial activity in response to a stimulus; then possess two or more killing mechanisms [151,152].

4.1. *Use of metals, metal oxides and metal nanoparticles (NPs)*

Ever since prehistoric times, metals like copper and silver have been used as antimicrobial materials, but their biocidal activity has remained incoherent [20]. Several studies have specified that different metals/alloys affect microbes differently due to the oxidative stress, protein disfunction or membrane impairment [153]. In the past decades, numerous metal ions and their oxides have been studied for their antimicrobial activity, particularly silver, copper, and zinc ions/oxides. Additionally, these metals/alloys displays lower toxicological effect and high antimicrobial activity making them suitable for antimicrobial applications [154]. The rudimentary chemistry behind the working principle of metals can be considered as (i) Hard–soft acid base theory (HSAB theory) which has been delineated observation, conveys an ordering of transition metals according to their preferences for certain organic ligands (proteins) [20]; (ii) reduction potential-ability to participate in redox reaction. In general, copper, silver, titanium, zinc, zirconia etc. are the most used and are lethal to bacterial cell wall, however other metals such as iron, chromium, mercury, tellurium, nickel, cobalt, etc. are either associated with some organic materials or polymers before being used as antimicrobial agents [155].

Nanotechnology offers a sound manifesto for adjusting the physicochemical properties of numerous materials to create efficient antimicrobials [156,157]. Metallic nanoparticles could be advantageous as their surface area is exceptionally large relative to their size, hence their biocidal efficiency is better [158]. Few others have also reported that due to crystallographic surface structure, there is an increase in the bactericidal activity of NPs [159]. Many researchers have described that since the mode of action in NPs is primarily by direct contact with the bacterial cell wall, the problem of growth of antimicrobial resistance (AMR) by bacteria can be amended [160]. Various metal and metal oxide nanoparticles (NPs) such as silver, gold, titanium dioxides (TiO_2), zinc oxide (ZnO), and magnesium oxide (MgO), exhibit a wide range of biocidal activity against both Gram-positive and Gram-negative bacteria [161,162]. Three possible mechanisms have been reported to describe the antimicrobial activity of metallic NPs:

(i) Disruption of bacterial membrane through the physical structure of nanoparticles, [99];
(ii) Production of oxidative stress by the development of reactive oxygen species (ROS), [68] and
(iii) Enhanced discharge of free metal ions from the NPs surfaces [156].

Many researchers have also reported the effectiveness of the synergistic antibacterial activity of these NPs when used in combination with other antibacterial agents such as polymers and metals [76,118]. The potency of each different metals, metal oxides and their NPs are discussed here.

4.1.1. Copper (Cu)

Many researchers believe that antimicrobial activity triggered by the Cu ions is the finest solution to kill microbes rapidly [111,163]. Most of the researchers have identified that Cu surfaces kill microbes through contact killing [164]. It is understood that loss of microbial cells has been a result of increased reactive oxygen species (ROS) caused by the Cu ions. It can be hypothesized that this leads to protein peroxidation, loss of membrane integrity and finally cellular death [165]. Recently researchers have reported that among all the material surfaces, the novel coronavirus (SARS-CoV-2) was decayed in about 4 h on a Cu surface and after 1 h on the cuprous oxide (Cu_2O) coated surface [166]. Fujimori *et al.,* [167] proposed two possible mechanisms involved in the inhibition of microbes on a Cu surface. The first method explains occurrence due to the release of reactive oxygen species (ROS) which can be shown by the following equations:

$$2Cu^{+}+2O_2(aq.)\rightarrow 2Cu^{2+}+2O_2{}^{-} \tag{1}$$

$$2\,O_2{}^{-}+2H^{+}\rightarrow H_2O_2+O_2\ \text{(Haber − Weiss reaction)} \tag{2}$$

$$Cu^{+}+H_2O_2\rightarrow Cu^{2+}+OH^{-}+OH\qquad \text{(Fenton reaction)} \tag{3}$$

It may be inferred that the hydroxyl radicals generated during these reactions damage the cell wall of the microbes.

A second mechanism suggests that Cu (I) and Cu (II) react with lipids of the microbes and result in the formation of lipid peroxide which destroys the DNA inside the microbes [167]:

$$Cu^{+}+LOOH\rightarrow Cu^{2+}+LO+OH^{-} \tag{4}$$

$$Cu^{2+}+LOOH\rightarrow LOO.+H^{+} \tag{5}$$

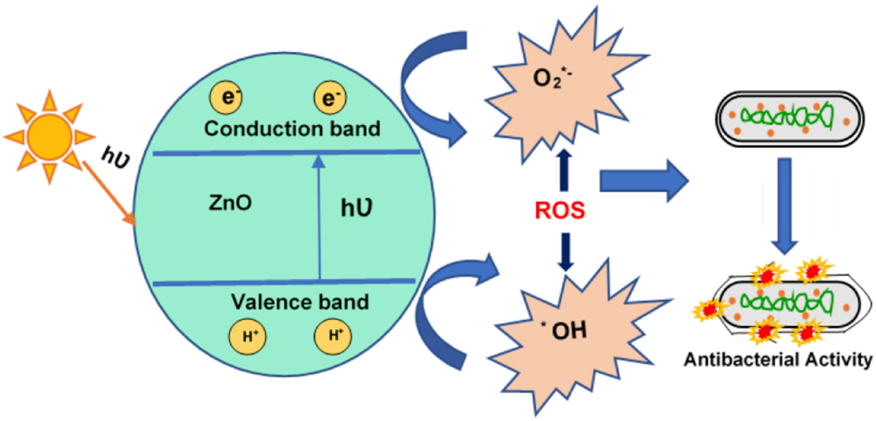
It was also reported that the production of Cu ions as per the second method is more efficient in eradicating microbes and viruses than the first method [168]. Recently, Cu ions were impregnated on a metal surface along with polymers to enhance the capability of killing two or more microbes at the same time. Such advancement in using Cu-Ag ionization in water was found to be effective in destroying a microbe such as *Legionella* spp. in contaminated water supplies for the hospitals [169]. Similarly, it was also reported that Cu (II) exerts antibacterial synergy with QACs which is used in preserving wooden surfaces from *Pseudomonas aeruginosa* [170]. Therefore, utilisation of copper has been an attractive option in many applications especially in hospitals, however, in normal places such as houses, and offices, it is avoided due to aesthetics.

Copper NPs are very effective against bacterial, fungal and viral infections [171]. Copper oxide (CuO) NP shows antibacterial activity through ROS formation and membrane destruction [165]. Other researchers have also reported that when contact is disrupted, Cu NPs can undergo lipid peroxidation, damaging the DNA of bacterial cells [172]. Many studies have shown that copper NPs kill microbes within a few minutes due to the contact kill mechanism [172]. However, owing to their small sizes, Cu NPs cause cytotoxicity and genotoxicity and, this must be considered before use [108]. Copper oxide (CuO) NPs are effective against a variety of microorganisms, but their antibacterial activity is slightly lower than that of Ag or ZnO. Therefore, unlike other NPs, CuO NPs would need a higher concentration to achieve the same antibacterial performance [111]. In addition, exposure to air rapidly oxidizes copper NP, limiting its application [161]. In essence, it may be inferred that the use of Cu NPs alone is not effective against microbes. It is advisable to use Cu NPs in combination with other antimicrobial agents.

Construction application: Jedrzejczak *et al.,* [173] pointed out that cement samples with CuO (0.5 %) exhibit the most inhibitory effect compared to ZnO against *S. aureus, E. coli and P. aeruginosa.* In many literatures, it has been reported that the use of copper plated surfaces in hygienic areas such as the hospitals are recommended as it can retain the bactericidal efficiency for more days [111]. Consequently, copper alloy surfaces could be employed in communal areas and at any mass gatherings to help reduce transmission of respiratory viruses from contaminated surfaces and protect public health [168]. Wrona, *et al.*, [174] conducted experiments on protective copper-based coatings which were deposited on stainless steel substrates by plasma spraying. They found that within 2 h, the deposited surface showed antimicrobial activity against *E. coli whereas S. aureus and P. aeruginosa* showed some effects in 24 h. However, their efficacy was reduced when deposited on polished surfaces. Addition of copper phthalocyanine was found to improve workability and strength of concrete and parallelly it was found that the sterilizing rate of copper phthalocyanine on Bacteroidetes and Proteobacteria, which were the main microorganisms causing concrete corrosion, were the highest (90.82 % and 64.25 %) respectively [175]. Latex paints containing copper-glass ceramic powder showed $\geq$ 99.9 % reduction in S. *aureus, P. aeruginosa, K. aerogenes and E. Coli* [176]. Many authors found the beneficial use of copper ion in geopolymer mortar or geopolymer concrete. It was inferred that geopolymer ion-exchanged with copper ions suppressed the growth of fungi such as oyster mushrooms in sawtooth oak sawdust, indicating antimicrobial activity [58]. Despite advantages, Paton *et al.*, [177] reported that pure copper and its oxides (which naturally form in air) can corrode over time leading to loss of function.

4.1.2. Silver (Ag)

The antibacterial action of silver is utilised and known in numerous applications for more than two decades [178]. Ag ($\overset{o}{E}=+0.80$ V) releases ions which makes it antibacterial. Owing to high price for pure silver, it is generally deposited electrolytically by doping with polymers [179]. It has been observed that Ag ions intermingle rapidly with the thiol groups of enzymes of bacteria that is responsible of its replication. Hence, Ag ions interrupt the process of replication and ultimately kills the microbes [164]. Apart from high price, one problem with silver is that the oxidation process of Ag^{+} ions under normal conditions is slow. It has also been



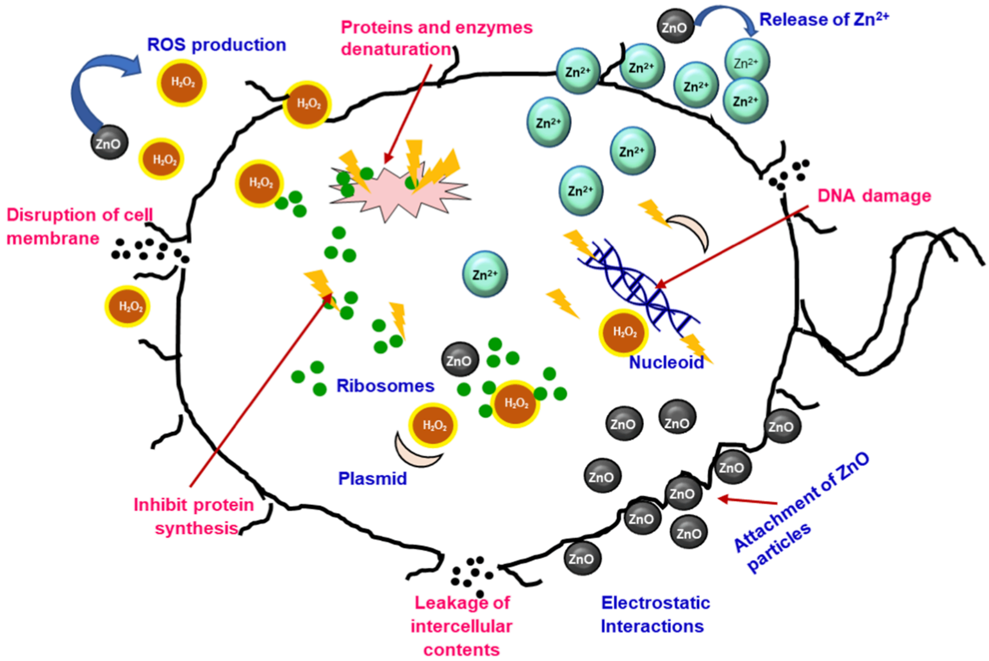


Fig. 12. Biocidal mechanism of ZnO: (a) Photocatalytic; (b) electrostatic interaction and release of Zn^{2+} ions.

highlighted that the Ag coating deposited on a rougher surface is not effective in killing microbes, especially *E. Coli* bacteria [180]. Therefore, it was found more effective to deposit Ag, when surfaces were coated with nanoparticles, salts of Ag, or polymers impregnated with Ag ions depending on the type of application [178,181].

Despite these inferences, silver NPs have been the most widely used metallic NPs as antimicrobial materials which work well against bacteria, fungi, and viruses in various industries [156,159]. It is noted that the Ag NPs caused "pits" in the bacterial cell wall by increasing the permeability of the membrane against *E. Coli* [156,182]. Other researchers have reported that Ag NPs can inhibit and destroy bacterial protein structures by binding to thiol and amino groups [116]. When combined with photocatalytic antibacterial materials such as ZnO and TiO_2, ROS can be produced, causing cells to immediately explode and kill bacteria [178]. All in all, coatings comprising Ag may prove to be a real asset for any applications in decreasing the number of microbial adhesions. However, the nature of the silver particles as well as how these are incorporated in the coating will determine the efficacy of surfaces coated with silver or silver nanoparticles. Ag NPs were found to exert an antifungal activity when evaluated against *C. albicans* by disrupting the structure of the cell membrane and inhibiting the normal budding process due to the destruction of the membrane integrity [183].

Construction application: Wide range of literature reports the benefits of adding Ag or Ag NPs into the construction materials

Table 1
Concentration of Ag^{+} and Zn^{2+} (ppm) release from AgCl/ZnO coated stones [184].

	1d	10d	20d	30d	40d	50d	60d
Ag^{+}	3.5	10.7	10.3	10.1	10.3	10.3	10.4
Zn^{2+}	55.6	82.5	82.8	82.7	83.4	82.9	83.9

resulting in better antibacterial activity. Mu *et al.*, [184] studied the effect of addition of AgCl nano-composite protective coating on stone-made building materials and found that antimicrobial activity remains stable even after prolonged exposure to the natural environment (60-days). Nam [185] reported that although the initial and setting time increased with impregnation of Ag NPs in the Portland cement, 5 % addition of Ag NPs by weight into the cement proved to perform better antibacterial material. This effect was prominent especially against *Candida albicans*, proving it to be an antifungal material [183]. Additionally, Paiva, *et al.*, [186] stated that addition of Ag NPs in cement improved 32 % compressive strength when measured after 28 days. Adak *et al.,* [187] doped silver NPs into alkali-activated mortar and found that antibacterial properties improved due to the inhibition of silver ions on bacterial enzyme activity and the destruction of cell permeability. It was also noted that low calcium fly ash-based geopolymer concrete containing a few oxidized Ag ions/NPs gets attached to the bacterial surface electrostatically and thus reduced the osmotic stability of the bacterial cell, trailed by consequent leakage of intracellular constituents of the bacteria [188]. Tsai *et al.*, [189] investigated the biocidal activity of silver-coated Au nanoparticles that are immobilized on cellulose paper. Such paper coated with 15 nm Au-Ag nanoparticles was observed to have excellent antibacterial activity against *E. coli.* Many researchers believe that the addition of silver in hybrid with other nanocomposites will improve both durability and efficiency of the material and prevent toxification to humans [190].

4.1.3. Zinc (Zn)

Zinc (Zn) and Zinc oxide (ZnO) are promising antimicrobial metallic agents used in different applications in medical industries [191]. It has been hypothesised that the mechanism of antimicrobial activity of ZnO is based on the release of burst of Zn^{2+} ions, albeit, ZnO nanoparticle-induced oxidative stress can trigger apoptosis (cellular death) too [192]. Kumar *et al.*, [193] explained that ZnO can interact with a bacterium both chemically and physically. Chemically, ZnO can react with the bacterial cell resulting in a photo-induced production of ROS with the formation and release of H_2O_2 and Zn^{2+} ions. These products damage the cell membrane of bacteria and DNA leading to cell death [191,194]. The physical interaction is mainly the result of cell envelope rupture and mechanical damage caused by ZnO with bacteria.

An illustrative explanation about the mechanism of chemical interaction between ZnO and bacteria is shown in Fig. 12. It was suggested that the positively charged ZnO undergoes photo-induced molecular transformation in the presence of UV-light (<385 nm). It causes a redox reaction resulting in the generation of free radicals such as hydroxyl (OH^-), O_2^-, H_2O_2, etc. responsible for damaging DNA of the bacteria cell [195]. Studies have revealed that ZnO coating exhibits better antimicrobial activities against gram positive bacteria (*B. Subtilis, S. aureus*) than the gram negative bacteria (*E. Coli*) [196]. It is theorised that the existence of an extra outer plasma membrane with thick lipopolysaccharide layer that oppose lipid peroxidation in presence of ROS makes it more resistant to ZnO coatings [197]. Few studies have reported that opposite charge of bacterial cell and ZnO particles are responsible for strong electrostatic attraction between them resulting in increased surface tension and membrane depolarisation [198,199]. Thus, it leads to membrane rupture causing cell death. A potential problem with zinc coating is that it is likely that during coating, metal may interact with hydrogen molecules. This can cause hydrogen embrittlement leading to weakening and cracking of the surface which can be improved by doping with zinc phosphate [192]. Despite various advantages offered by the ZnO coatings, it is also observed that the use of ZnO in high concentration against S. mutants, toxicological impact and undesirable effect on human body may prevail [200]. However, Carvalho *et al.*, [201] suggested that an increased thickness of the ZnO coating to 200 nm performs more effectively against *E. Coli* compared to other thickness. ZnO doped with acrylic paints were found to be more suitable to use for inhibition of fungal growth in indoor environments such as kitchens, bathrooms, sinks, etc [9].

Construction application: ZnO nanoparticles were identified to exhibit antimicrobial activity against several microbial species. Many researchers believed that the ZnO nanoparticles coatings enhances the durability of stone surfaces against fungal attacks (*Aspergillus Niger* and *Penicillium sp.*) due to ROS production in the presence of UV light [35]. However, Ruffolo and La Russa [36] explored the feasibility of effectiveness of ZnO and $ZnTiO_3$ nano-coatings on stone heritage samples and noted that higher inhibition was observed in $ZnTiO_3$ than ZnO even though both are hydrophobic and photocatalytic. The application of Zn-doped MgO NPs obtained by the sol − gel method as antifungal coatings on dolomitic and calcite stones has been explored to develop effective protective coatings for stone against *Aspergillus niger, Penicillium oxalicum, Paraconiothyrium sp.*, and *Pestalotiopsis maculans*, which are active in bio-weathering of stone [202]. Mu *et al.,* [184] studied the effect of addition of AgCl/ZnO nano-composite protective coating on stone-made building materials and found that antimicrobial activity remains stable even after prolonged exposure to the natural environment (60-days). Table 1 shows the concentration of Ag^{+} and Zn^{2+} release which was observed to inhibit microbial/fungal growth.

The improved photocatalytic and antifungal properties detected in Zn-doped MgO NPs was attributed to the formation of crystal defects by the incorporation of Zn into MgO. A few studies have explored the possibility of using ZnO, Ag NPs or combined ZnO/Ag surface modified bricks against *S. aureus* and *Bacillus cereus*. They reported that production of ROS in the presence of sunlight made the bricks self-bactericidal material [203]. Singh *et al.*, [204] observed the positive influence of adding ZnO in cement composite against bacteria (*E. Coli and B. Subtilis*) and fungal growth (*A. Niger*). Bacterial growth was reduced with increase in ZnO concentrations from 5

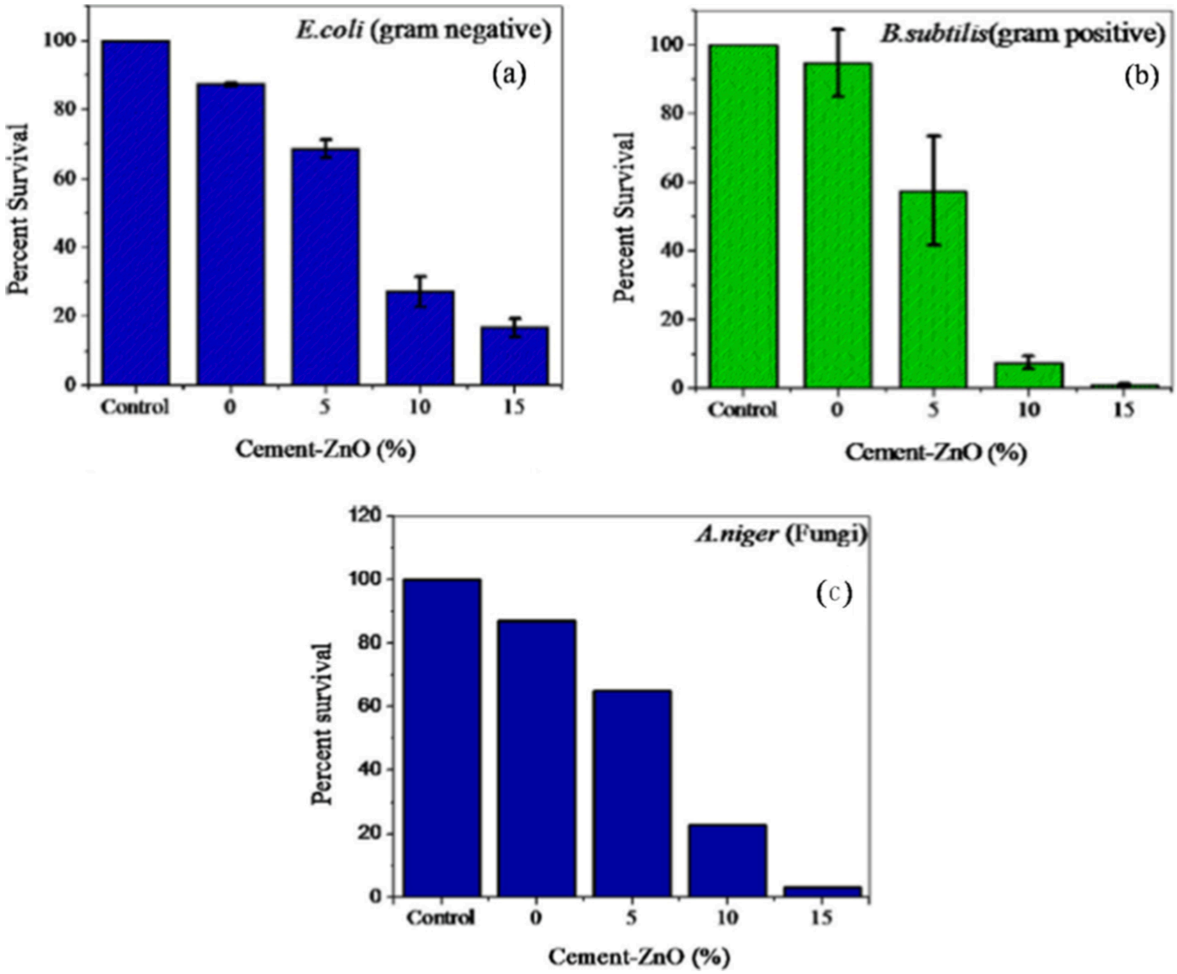


Fig. 13. Effect of ZnO (%) in cement composites under dark (a) *E. Coli*; (b) *B. Subtilis* and (c) *A. Niger* [204].

wt% to 15 wt% under dark (Fig. 13). It was seen more effective in case of gram-positive bacteria (*B. Subtilis*) than the gram-negative bacteria (*E. Coli*). Similar results were found for *A. Niger*. Additionally, it was also reported that antibacterial and antifungal activity improved in the presence of solar light than dark.

Contradictorily, Jedrzejczak *et al.,* [173] reported that 0.1 % of ZnO in cement composites showed higher efficiency against *S. aureus* and *E. coli*. The photocatalytic and hydrophobic nature of nano ZnO embedded in concrete mitigates *thiooxidants* and *thiobacillus* prevented microbiological induced corrosion [204]. Likewise, ZnO NPs were also found to be more effective in case of paper products. Ghule *et al.*, [71] characterised and investigated the efficiency of ZnO NPs on the surface of paper made from cellulose fibers. It was found that the ZnO NP coated paper showed excellent biocidal activity against *E. coli* for 24 h due to the production of H_2O_2. Likewise, Jaisai and Dutta [70] developed an antimicrobial paper by growing ZnO nanorods and testing it against *S. aureus, E. Coli* and *Aspergillus niger*. It was concluded that the formation of H_2O_2 and O_2^- products in the presence of light initiated the deterioration of paper samples. Several researchers have developed transparent glasses with a high content of ZnO to show excellent antimicrobial activity against *E. Coli, S. aureus* and yeast [205]. The release of Zn^{2+} ion increases the cytotoxicity with cell viability greater than 80 %.

4.1.4. Titanium dioxide (TiO_2)

Titanium (Ti) and titanium dioxide (TiO_2) have been widely used as orthopaedic and odonatological implant materials due to their suitable biocompatibility, outstanding corrosion resistance, light weight and high mechanical strength [206]. TiO_2 is also known to be a photo-catalyser element and is used in self-cleaning tiles, glasses, windows etc. [207]. Being a photo-catalytic material, the antimicrobial activity of TiO_2 is activated on being irradiated with UV radiations (<385 nm) [207,208]. Researchers have noted that surfaces coated with TiO_2 prevents initial adhesion of microbes due to the production of ROS [209–211]. The main advantage of using photocatalytic surface is that no external electrical power or chemical reagents are required to perform their function and it only require source of light, oxygen and sometimes water [212]. The basic mechanism with which TiO_2 catalyst surface operates can be described by the following equations:

$$TiO_2 + h\upsilon \underset{(catalyst)}{\rightarrow} e^- + h^+ \tag{6}$$

$$h^+ + H_2O \rightarrow HO^* + h^+ \tag{7}$$

$$h^+ + OH^- \rightarrow HO^*(\text{surface}) \tag{8}$$

$$e^- + O_2 \rightarrow O_2^* \tag{9}$$

$$2O_2^* + 2H_2O \rightarrow 2HO^* + 2OH + O_2 \tag{10}$$

It has also been reported that TiO_2 acts as a depolluting agent that can be deposited on concrete pavement surfaces and external building surfaces which in the presence of sun light, removed the gaseous pollutants before these get washed away by rain [207,213]. The settled wisdom here is that the production of ROS products such as HO^*, O_2^* leads to degradation of microbial cells owing to oxidation of lipids and membrane disintegration. However, it is still unclear in the literature as to which radical is responsible for microbial death [209]. Recently, titanium doped with copper, nitrogen or silver was found to have more promising activity than pristine TiO_2 coating as it is suitable to kill two or more different microbes at the same time [206,208,214]. One of the demerits of TiO_2 coatings is that they do not denature microbes fast enough, even though it directly attacks the DNA of the bacteria. To improve this aspect, Yusuf *et al.*, [215] found that laser textured TiO_2 ceramic coatings possess improved and reliable antimicrobial performance.

TiO_2 is emerging as an important antimicrobial NP [156] due to the fact that it is photocatalytic i.e., its toxicity gets triggered by the UV light that causes ROS. ROS damages the membrane and DNA of the cell wall of the bacteria [160]. A versatile function of TiO_2 is that it can simultaneously serve as a photocatalytic antimicrobial material as well as a structural material which makes it suitable for use in the exterior and interior construction for purposes of making cement mortar, exterior tiles, sidewalk blocks, glass, and PVC fabric [207]. Photocatalytic building materials based on TiO_2 nanoparticles are mainly used to control environmental pollution for self-cleaning and self-disinfection. In combination with other metals such as silver and copper, TiO_2 nanoparticles are used in various areas such as water treatment, painting, etc., as excellent biocides against *Staphylococcus* and *E. Coli*, etc.

Construction application: Becerra, *et al.,* [216] found a synergistic effect of Ag-TiO_2 based nanocomposites stabilized by citrate, and achieved enhanced biocidal effect while maintaining colour alterations at a low level. TiO_2 is a widely used coating on wood preventing the growth of fungi and cyanobacteria. Goffredo *et al.,* [217] and Jnido, *et al.,* [218] evaluated the efficiency of an isolated solution of 1 % TiO_2 NPs as well as by combining it with Ag and Cu, which turned out to be more effective in both pine (softwood) and beech (hardwood) against *Aspergillus niger*. Similarly, a few studies have reported that fungal infestation in wood is very common, for which photocatalytic oxidation by TiO_2 has shown to deactivate the growth of *Aspergillus niger* [219]. TiO_2 modified building materials are most popular because TiO_2 has been traditionally used as a white pigment [207]. The major applications of TiO_2 based photocatalytic building materials include environmental pollution remediation, self-cleaning and self-disinfecting. The advantage of using solar light and rainwater as a driving force has opened a new domain for environmentally friendly building materials [131,207]. Vishwakarma *et al.*, [220] reported that TiO_2 NPs modified fly ash mortar and NPs modified $TiO_2 + CaCO_3$ fly ash mortar showed its efficacy in antibacterial activities against *Pseudomonas, Fusarium, algae,* blue-green algae, and manganese oxidizing bacteria. Likewise, Jedrzejczak *et al.,* [173] found that addition of 1 % of TiO_2 in cement inhibited the growth of *B. cereus* and *P. aeruginosa.* Vishwakarma *et al.,* [220] determined that addition of 2 % TiO_2 in fly ash mortar improved antibacterial activity by lowering the pH, and oxidising sulfur add further to inhibit bacterial growth. The hydraulic lime mortars mixed with porous microspheres of TiO_2 demonstrated the best antifungal activity [221]. Likewise, Jerónimo, *et al.*, [221] observed that addition of nano TiO_2 in hydraulic lime mortar causes significant differences in properties such as workability, compressive and flexural strength. This problem was further solved by varying the dosages of superplasticiser addition. Kumar, *et al.,* [222] investigated the durability performance of concrete which incorporated different dosages of TiO_2 and rice husk ash, and inferred that 10 % RHA and 3 % nano TiO_2 as a partial replacement of cement showed the highest strengths and durability performance alongside better antifungal properties. TiO_2 based paint is one of the well-known applications carried out by different researchers on steel, and concrete walls [223,224]. Researchers witnessed inhibition of growth of fungi viz. *Trichoderma viride, Aspergillus niger, Coonemeria crustacea, Eurotium herbariorum, and Dactylomyces sp*. which is due to the photocatalytic reactions [225]. Jin *et al.*, [226] evaluated the efficiency of Ti-Cu on SS316L stainless steel substrates and found that the formation of a Ti-O passive layer on the surface was responsible for inhibiting the sulphur reducing bacteria and *E. Coli*. Won, *et al.,* [227], prepared TiO_2-based transparent coatings on glass surfaces and found that the self-cleaning potential property of the glass inhibited the growth of fungi. One of the major demerits of the use of TiO_2-based incorporation or coating is that the photocatalytic ability of TiO_2 gets deactivated over time. Numerous studies have been undergone to recover its ability by thermal regeneration, flowing humid air over the catalyst, O_3 purge in the presence of water vapour and washing with alkaline solutions [228].

4.1.5. Molybdenum trioxide (MoO_3)

Molybdenum trioxide (MoO_3) is a transition metal oxide with *n*-type semiconductor which has become one of the most promising antimicrobial materials [229]. It is also known for its multifunctional properties like photocatalyst, photochromism, oxidation catalyst, and supercapacitors [230]. Various processes have been developed to produce these transition metal oxides, of which electrochemical deposition is the most widely used process [231,232]. From several studies it was found that due to the small size and high specific surface area, the electromagnetic interaction between the positively charged MoO_3-NPs and the negatively charged bacterial cell [230,233] were responsible for bactericidal activity of MoO_3. According to many researchers, the larger surface area of MoO_3 NPs

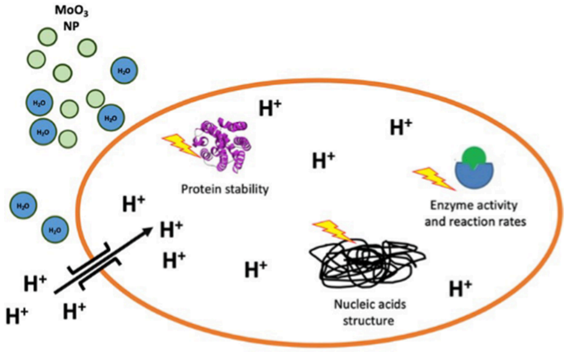


Fig. 14. Schematic diagram illustrating the bactericidal activity of MoO_3 [232].

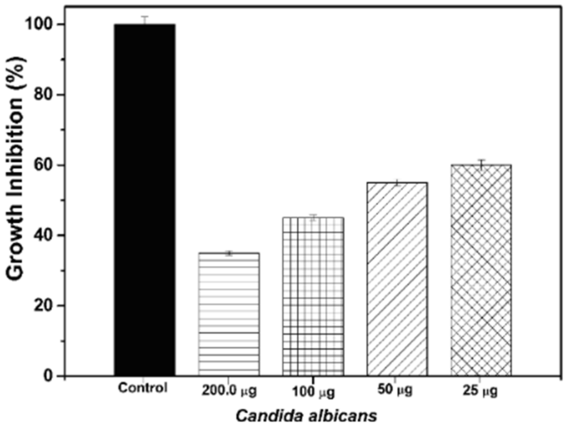


Fig. 15. Effect of addition of nano MoO_3 on bricks against *C. albicans* [239].

allowed them to bind tightly to microbes and destroy them through the release of metal ions [234]. A few other researchers have reported that the release of hydronium ions is related to the bactericidal activity of MoO_3, which affects the stability of proteins and nucleic acids, which in turn kill microbes [232] as shown in Fig. 14.

$$MoO_3 + H_2O \rightarrow H_2MoO_4 \tag{11}$$

$$H_2MoO_4 + H_2O \rightarrow H_3O^{+} release \tag{12}$$

It is known that inhibition of gram-negative bacteria is more difficult with NPs than the gram-positive bacteria due to the permeability of cell wall [235]. Due to the higher inhibition zone, MoO_3 NPs interact much better with gram-negative bacteria than other NPs and at the same time kill them immediately. A few other researchers have argued that MoO_3 possess large band gap and photocatalytic properties which improves the antibacterial activity against *Candida albisane and Aspergillus Niger* [233]. In addition, MoO_3 NPs are also observed to work and have proven their exceptional effectiveness in killing many types of microbial species and in combating antibiotic resistance (AMR) [236], especially when doped with photocatalytic metal oxide ions [237].

Construction application: Yin *et al.,* [238] investigated the synergistic effect of plasmonic MoO_{3}-x nanosheets doped with Ag nano-cubes and tested against near infrared light (NIR). The three main observations on microbial killing they noted were (a) Bacteria gets killed due to near infrared absorption and photothermal conversion; (b) Heat induced in MoO_3 by near-infrared radiation causes Ag nano-cubes to release Ag^{+} ions that kill bacteria; and finally (c) the formation of ROS at the MoO_3-Ag interface due to the transfer of hot electrons into the band gap oxidizes the bacterial cell wall killing them instantaneously [238]. Zollfrank *et al.*, [236] explained that MoO_3 sol–gel based coating can kill *staphylococci, streptococci, enterococci, Legionella pneumophila, Lactobacillus acidophilus* spp.*, Candida* spp.*, Aspergillus* spp. based microorganisms under pH 3.5–5 and killed *E. coli, Pseudomonas aeruginosa, Clostridia, Campylobacter* when pH > 5.5. Alghamdi, [239] synthesised MoO_3 nanobricks using a hydrothermal method and conducted antimicrobial tests against *S. epidermidis, S. aureus, E. Coli* and *C. albicans* as per ATCC 29213. It was observed that MoO_3 generated an acidic medium which inhibited the growth of microorganisms (Fig. 15). MoO_3 paint coated surface exhibit a significant loss of viability in a time-dependent manner especially against Escherichia coli, *Pseudomonas aeruginosa, Staphylococcus aureus, and Klebsiella pneumoniae* [240]. Combined composites containing MoO_3-SiO_2-Ag_2O based coating possessed higher antibacterial activities than the MoO_3-SiO_2 coating, due to the combined effects of surface hydrophobicity, the release of Ag^{+} ions, surface acidic reaction and photocatalytic

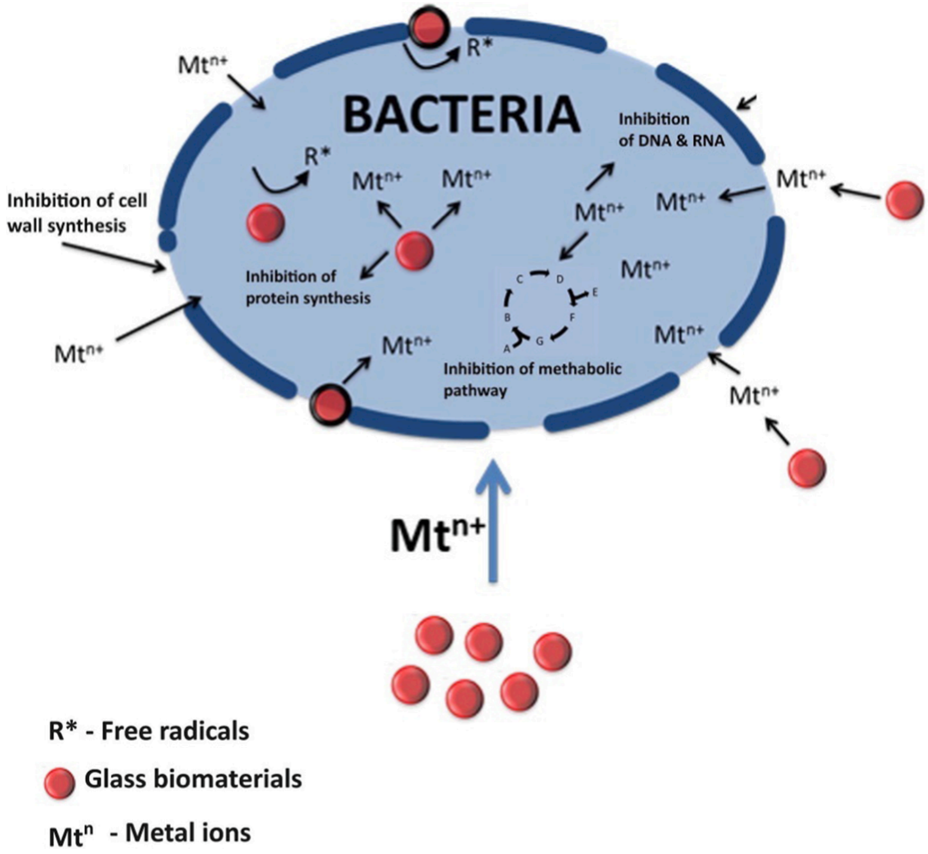


Fig. 16. Antimicrobial activity of bioactive glasses [247].

activity [241]. Such composites result in fabricating self-disinfecting surfaces and can be used in reduction of hospital-acquired bacterial contaminations.

4.1.6. Other metallic nanoparticles

A few other metallic NPs that are used in different applications are silica, Fe_3O_4, Au, MgO, Al_2O_3 etc. These materials are mainly used in the medical field, especially in making antibiotics [156]. It has been observed that the bactericidal mechanism of magnesium oxide (MgO) nanoparticles is related to the production of high concentrations of superoxide anions ($-O_2$) on MgO surfaces, which can react with the carbonyl groups of peptide linkages in bacterial cell walls to destroy them [182]. Additionally, it has been reported that gram-negative bacteria are more susceptible to Mg ions than gram-positive bacteria due to differences in their membrane structure Sikora *et al.*, [242] studied four nano oxides (Al_2O_3, CuO, Fe_3O_4, and ZnO) added in composites against different bacterial species and concluded that metal oxide NPs may not be efficient in preventing microbial growth when dispersed improperly. Au NPs are considered to be so valuable in the development of antibacterial agents due to their nontoxicity, high ability to functionalization, polyvalent effects, ease of detection and photothermal activity [161]. Bactericidal activity of the Au NPs was attributable to attachment of these NPs to the bacteria membrane followed by membrane potential modification and ATP level decrease [243].

Construction applications: The application of MgO- and Zn-doped MgO NPs as protective coatings on calcareous stones showed important antifungal properties, inhibiting successfully the epilithic and endolithic colonization of *A. niger* and *P. oxalicum* in both lithotypes, and indicating a greater antifungal effectiveness on Zn-doped MgO NPs [202]. Merachtsaki, *et al.,* [244] explored the possibilities of addition of MgO based coating against corrosion resistance and found that MgO based coating can easily resist the growth of sulphur reducing bacteria, preventing corrosion due to sulphur attack. Interestingly, some researchers have made calcium-based cement using eggshells and reported that cement's biocidal activity is effective and more specific for *Streptococcus mutants and E. faecalis* [245]. One of the literature sources reported that adding four different nanoparticles of nanometal oxides (Al_2O_3, CuO, Fe_3O_4, and ZnO) to Portland cement improved the mechanical properties and durability of cement and cement-based composites, but it was not effective in preventing microbial growth when it is randomly distributed in the matrix [246]. Bioactive glass applications have become increasingly popular in the medical field and in the construction industry [247]. Bioactive glasses are those in which anions (Cl^-, F^-) and cations (Ag^{2+}, Cu^{2+}, Na^+, Ti^{2+}, etc.) are added to the silica matrix during production of glass products [75]. As shown in Fig. 16, bioactive glasses act as conductors for the local release of metal ions that inhibit the growth of microorganisms [247]. Cacciotti [248] developed bivalent cationic ion doped bioactive glasses where Mg, Zn, Sr and Cu ions were doped in combination. The synergistic effect was detected with CuO-ZnO, Zn-Mg and Sr-Mg, especially due to the release of the ions that degrade the bacterial cell.

4.1.7. Zeolites

Zeolites are nano porous aluminium oxide silicates made of silicon, aluminium and oxygen in pores along the frame. Zeolites are an ideal material to host and release the metal ions because of their controllable ion exchange properties and the high thermal and chemical stabilities desired for industrial production processes [249]. This makes them desirable to kill any type of bacterial strains. Their cation contents can be exchanged with monovalent or divalent ions [250]. The entire structure of zeolite has negative charge, and this charge is balanced by ions of other atoms such as Na^{+}, Ag^{+}, Zn^{2+} and Cu^{2+} etc. Cation exchange, especially silver exchange, is one of the properties of zeolites that give them prolonged antimicrobial properties against *E. coli, Bacillus subtilis*, and *S. aureus* [251]. Additionally, some researchers believed that silver ions are highly polarizable due to the strong electric fields within zeolites and are very tightly bound to the anionic framework, creating a highly durable surface with very little leaching [177]. Ag-zeolite demonstrates superhydrophilicity, ensuring contact between the silver ions and suspended microorganisms; studies have demonstrated high antimicrobial activity against *E. coli* when submerged in deionised water for long periods and through repeated bacterial exposures [177].

In addition, other heavy metals such as zinc, copper, nickel, mercury, tin, lead, bismuth, cadmium, chromium and thallium, have also been found to be effective in inhibiting bacterial growth [250]. Interestingly, Torres-Giner *et al.*, [252] tried a multi-ionic Ag-Cu-Zn zeolite and found that it was the most efficient antimicrobial sample. Several studies have investigated the effectiveness of zeolite / TiO_2 hybrid composites and found that they have good photocatalytic degradation performance [253]. The synergistic effects of zeolite and TiO_2 produces ROS and involve high oxidative stress in killing germs. However, the use of zeolites is still limited in many industries, which is likely due to the existence of several controversial antibacterial principles and several factors [250].

Construction applications: For more than twenty years, zeolites are being used to achieve bactericidal effect through the release of metal ions (Ag, Cu, and Zn) by their addition into concrete [19]. Many researchers have reported that the release of Cu^{2+} and Zn^{2+} from the zeolites are the main cause of DNA damage in bacteria and denature of proteins and nuclei [56,254]. Several studies have established that the use of silver-copper ion zeolites performs biocidal activity exceptionally well against *E. Coli, S. aureus and Salmonella enterica* when their concentration is kept more than 3 % incorporated in cement mortar [223,224]. It was also described that the release of co-cations such as Cu^{2+} and Zn^{2+} are responsible for antimicrobial activity of zeolites. Addition of zeolites (5 %) into concrete was observed to have a slight decrease in the mechanical properties, however, it showed enhanced biocidal activity against all types of gram-negative or gram-positive bacteria [19]. Sewer concrete containing Ag-zeolites was found to be effective in killing *Acidthiobacillus* and *thiooxidants* thus resulting in prevention of microbiological induced corrosion [254]. Qing, *et al.*, [255] used porous 316 L stainless steels scaffolds which were fabricated by selective laser melting technology with a zeolite coating and tested against *E. coli* and *S. aureus* bacteria. They were seen to inhibit *E. coli* and *S. aureus* after 24 h of incubation. Paton, *et al.,* [177] reported that the superhydrophilicity nature of Ag-zeolites makes them suitable for manufacturing antimicrobial surfaces for use on inhabited space craft or other structures where light-weight, and high durability is very important.

4.2. Use of polymers

Antimicrobial polymers offer a promising strategy against microbes and have received attention in both academic and industrial research. Usage of antimicrobial polymers in various applications such as medical, food storage, and textile industries and in the construction sector as building materials and tiles etc. [22]. Polymers are claimed to have superior efficacy, reduced toxicity, and minimised environmental problem [256]. The antimicrobial activity of different polymers varies based on mode of action and type of monomers attached, the way they are applied and the surrounding environment [257]. Many researchers have suggested that control over hydrophobicity/hydrophilicity, cations, molecular weight, functional group, and degree of polymerisation may influence the bacterial activity [100,258] depending upon the type of the polymers as given below section.

4.2.1. Man-made polymers

The use of polymeric synthetic fibres such as polypropylene fibre, polyethylene fibre, poly (ethylene glycol) (PEG) and polyvinylalcohol fibre have found more rapid growth in recent years [259]. These man-made polymers are used to improve durability and strength and is gaining importance in the field of antimicrobial agent applications. These are either used individually or in combination of two or more polymers/metals/nanoparticles [2]. The modes of action of man-made polymers are classified as active and passive. A passive polymer can reduce proteins/lipids adsorption on its surface without killing the microbes, i.e. these polymers repel the microbes [260]. One of the known passive polymers is poly (ethylene glycol) (PEG) and it has been used due to its high chain mobility, chemical stability and steric hindrance effect [261]. Passive polymers are generally modified chemically or physically to enhance the antibacterial activity. It was highlighted that attaching one hydroxyl (OH^{-}) group in water-soluble PEG can result in excellent microbial repelling properties [91]. Likewise, many investigations have reported that an increase in density, grafting of polymeric surface, blending of another polymer, the addition of surface net charge monomer can reduce more than fivefold the adhesion of microorganisms [2].

Active polymers are functionalised with active agents like cationic biocides, peptides or antibiotics that can kill the microbes instantly [22]. Most of the polymers modified with quaternary ammonium compounds (QACs), quaternary phosphonium compounds (QPCs), metals ions and nanoparticles to enhance their antimicrobial capacity. These active polymers are further classified as polymeric biocides, biocidal polymers and biocide releasing polymers [14,262].

4.2.1.1. Polymeric biocides or antimicrobial polymers. These are those polymers which are covalently doped or copolymerised with a repeating group like amino, carboxyl or hydroxyl groups, i.e. the polymers are just multiple interconnected biocides, which act similarly to monomers [22]. Habitually, the polymerization of biocidal monomers does not lead to active antimicrobial polymers, as they may be water-insoluble, or the biocidal functions do not strike their targeted microbes [101]. For instance, the polymerization of antimicrobial 4-vinyl-*N*-benzyl pyridinium chloride and subsequent crosslinking resulted in a non-biocidal water-insoluble polymer that only captures microbes but does not kill them [91]. These polymers typically include antibiotics/antibacterial agents blended with them, and thus majorly used in medical applications especially as a coating of urinary catheters, and surgical equipment [101]. The long-term inhibition (up to 9 months) of fungal infestation could be achieved through the application of specific biocides, namely 5-chloro-2-2methyl-2H-isothiazol-3-one (CMIT) and 2-methyl-2H-isothiazol-3-one (MIT) [9]. However, repeated maintenance is required to remove the fungal infestations.

4.2.1.2. Biocidal polymers. Biocidal polymers can kill microbes since they contain a cationic functional group or biocides that can destabilize bacterial cells leading to its death. The antimicrobial activity of the cationic group depends on charge density, pH and hydrophobic interactions [22]. Nowadays synthetic peptides with the amino group are the new generation of antimicrobial agents used to coat surfaces in hospitals. Timofeeva and Kleshcheva, [258] reported about the polycation antimicrobial function of biocidal polymers wherein cell death occurs due to electrostatic interactions between opposite charges. One of the known biocidal polymers namely poly(methacrylate), contains chlorhexidine-like side groups giving it a great potent against *S. aureus.* It is commercial and is used in many antimicrobial paintings and coatings [101]. Researchers have also observed that biocidal polymers work most effectively when cationic groups are positioned along the polymer backbone against all gram-positive and gram-negative bacteria [92,256]. Francolini *et al.*, [92] stated that with link of QACs biocides to polyurethane, biocidal activity enhances up to 90 % and this suppresses the growth of *S. aureus* and *E. coli* bacteria.

4.2.1.3. Biocide releasing polymers. These are utilised as biocide carriers such as gentamicin, triclosan, silver, chitosan, and nano-metals, which kills the bacteria on their interaction [22]. These polymers present excellent value due to the high concentration of biocide accomplished through the release and proximity to the target cells. However, the toxicity coupled with the biocide molecules is a severe disadvantage linked with this tactic, and their efficiency decreases substantially with time [14]. Synthetic biocide releasing polymers are highly scalable, cost effective and retain better chemical stability in a biological milieu [100]. Recently different types of polymers incorporating metallic nanofillers have been developed and used in diverse applications such as water treatment systems, and water storage systems [2]. These fillers are included in polymers via *in-situ* polymerisation or mechanical blending during their molten state [14]. The addition of antimicrobial agents to the polymers has indicated broad-spectrum antimicrobial activity, rapid bactericidal kinetics and a very low propensity to induce resistance and they are thus referred to as 'Biomimetic antimicrobial polymers' [88].

4.2.1.4. Zwitterionic polymers. Zwitterionic polymers such as polyampholytes refer to a group of materials which have equal number of cations and anions on polymeric chains [263]. In most cases, cations that are quaternized ammonium and zwitterionic group (sulfobetaine (SB) and caboxybetaine (CB)) as anions [264] are used as antimicrobial materials. It has been reported that due to the presence of many cations and anions, zwitterionic polymers possess super hydrophilicity and act as an antifouling agent. The anti-fouling activity of zwitterionic polymers is based on the strong interactions of these polymers with water through ionic salvation which is different than PEG [265]. Some recent investigations have reported that these polymers work under the principle of 'kill and release' strategy wherein bacteria is killed on the surface and dead strains are released upon hydrolysis of ether group in them [14]. Many researchers have also highlighted that zwitterionic polymers can resist non-specific proteins adsorption, bacterial adhesion, and biofilm formation [84]. Singha, *et al.,* [102] observed that zwitterionic polymers also form hydration layers but through tight electrostatic interactions unlike comparatively loose Van der Waals' force of hydrophilic coatings. This acts as a hindrance against foulants since the hydration layer does not let proteins settle down on the surface resulting in inhibition of bacterial adhesion. Recently, many studies have reported the advantages of polyzwitterionic surfaces, which are generally known as protein and cell-repellent materials, which repress amalgamation of microbes at the water interface [157]. Functionalisation using zwitterion polymers containing silica NPs inhibits protein (hydrophobins) adsorption and prevents fungal attachment. A reduction of spore attachment (below 100 counts) was observed within 24 h [266]. The structural versatility conferred to zwitterionic polymeric coatings due to its ability to attach various functional groups. For this reason, they are used in various applications such as in medical fields, drug delivery and marine [264].

Construction applications: Application of antimicrobial polymers are well documented in medical, food, and textile industries. Due to less knowledge about these polymers, their use in the construction sector is highly limited. Acrylic polymer coatings such as ethyl methacrylate (EMA), methyl acrylate (MA) and poly (methylmethacrylate) (PMMA), have been frequently used in stone conservation because they exploit both protective and consolidating properties. However, due to stability issues such as cross-linking reactions, acrylic polymers are rarely used as coatings on stone structures [267]. In recent years, wood-plastic compounds have been used mainly because of their low weight, low cost and flexibility in shape [268]. Manufacturing of polyvinyl chloride composite was found to be self-sufficient biocidal against *E. Coli* and *S. aureus*, Similarly, a few other researchers have remarked that cork wood has better antimicrobial properties against *E. Coli* and *S. aureus* [39].

Yu *et al.*, [269] observed that the addition of a silver NP coating on wood-plastic composites proved that it has good biocidal activity and improves the mechanical properties. Misra *et al.*, [270] observed that the use of polyoxometalate-ionic liquid coatings can

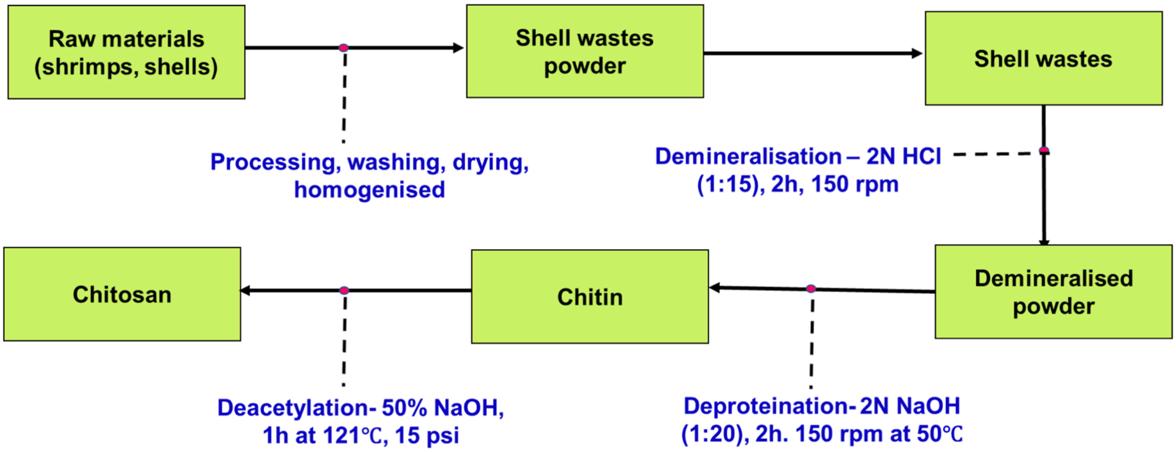


Fig. 17. Production of Chitosan from Chitin.

act as anticorrosion and antibacterial material for any type of natural stone against *E. Coli* bacteria. Conventional polymers being used to shield metal surfaces (steel, iron rods) from biocorrosion damage are mainly composed of polyurethane, fluorinated compounds, epoxy resins, polyimides, silicone, coal-tar epoxy, and polyvinyl chloride [271]. Videla *et al.,* [64], determined that polyvinyl chloride (PVC)-based coatings exhibit poor protection performance against corrosion induced by microorganisms. Many researchers have found that grafting polymers containing QAC covalently, immobilize a uniform monolayer of initiators on the metal substrate surfaces, that protects them against biocorrosion [22,41]. Water-soluble polyamine, polyethyleneimine polymers have been found to be more effective as a corrosion inhibitor to protect steel [272]. Several studies are being done to add a conductive polymer coating, i.e., addition of metal NPs by electrochemical reaction on a polymer substance were found to be effective against SRB causing corrosion. Superabsorbent hydrogel-silver nanocomposite based on poly(vinyl alcohol) showed very good antibacterial activity on gram-positive and gram-negative microorganisms when coated on steel surfaces [162]. Polymers such as poly(vinylidene fluoride) (PVDF), polyamide (PA), poly(vinyl chloride) (PVC), polypropylene (PP), poly(ethersulfone) (PES), polysulfone (PSf) and polyacrylonitrile (PAN), were used as a membrane layer in the structures which is used in water treatment plants, municipal waste management [82]. Owing to the desirable antifouling capacities, zwitterionic polymers have several applications, including biosensors, drug delivery, cell preservation, as well as chemical separation and marine coatings [264,273]. Zwitterion polymers such as sulfobetaine (SB), carboxybetaine (CB), phosphorylcho-line (PC) are also used as a membrane layer on the treatment plants. The membrane surfaces made with zwitterionic possessing low surface energy on their surfaces result in superior antifouling and self-cleaning properties [2]. Yuji *et al.,* [274] reported that zwitterionic poly(phosphobetaine) brushes and poly(sulfobetaine) brushes exhibited excellent anti-fouling characteristics for both macro- and micro-organisms (marine), whereas QAC brushes allowed mussel larvae settlement and bacteria adhesion. This result illustrates that potential of zwitterion polymer in the constructions which are more susceptible to biocorrosion. Copello *et al.*, [275] carried out studies on dodecyl-di(aminoethyl) glycine immobilised in silicon oxide xerogel matrix coated on glass against *E. Coli, Pseudomonas aeruginosa* and *S. aureus*. After 24 h, bacterial cells were damaged by a 99 % reduction in microbial colonization by xenografted glass and the reaction with xerogel was observed using SEM analysis.

4.2.2. Natural polymers

Natural polymers are biocompatible and biodegradable, and they can be obtained on a large scale and at a comparatively low price [14]. Until now, polysaccharides (Chitosan, cellulose), poly (ε-lysine), lignin, natural polyesters and peptides are used as antimicrobial agents in diverse applications such as medical, food storage and textile [78]. However, natural polymers are yet to be used in the construction sector. Some details about these nature polymers are discussed next:

4.2.2.1. Polysaccharides. These are macromolecules that contain proteins and nucleic acids [276]. It is recognised as a natural biodegradable antimicrobial material. Polysaccharides have many monosaccharides attached to them with varying groups such as amino or hydroxyl at the end of the chain and this leads to correspondingly varying properties [277]. Polysaccharides are now increasingly used in applications ranging from medical, food industries, and shipping containers. Previous studies have proved that the utilisation of polysaccharides in Portland cement retards the setting time and production of hydration products. Therefore, it can be understood that there is a high potential for using polysaccharides in Portland cement where handling and high temperature can be an issue.

4.2.2.2. Chitosan. Chitosan, amino polysaccharides have inherent antimicrobial properties. It is produced from chitin which is second most available natural polymer after cellulose [278]. Chitin is typically obtained from seafood industries such as shrimp and crustacean shells etc. [279]. Chitosan is produced from exhaustive alkaline deacetylation of Chitin. A simplified flow chart representing the production of Chitosan is shown in Fig. 17. Though there is a large number of studies revealing antimicrobial activity and application of Chitosan; the exact mechanism is yet understood [2]. Most researchers claimed that it originates from the electrostatic interactions between positively charged Chitosan and negatively charged bacteria [280–282]. A very few investigations have reported another mode of action raising from the formation of complexes with metal ions leading to disruption of DNA of the bacterium [283]. The antimicrobial activity of chitosan depends on pH, degree of deacetylation, ionic charge, cationic charge and density etc. [284]. The major application of Chitosan can be observed in medicals and food storage industries [285]. The effect of chitosan reduces in neutral pH conditions, its usage is however limited in other industries especially in the construction industry. Therefore, modification of chitosan by either attaching hydroxyl group, NH^{3+} group or doping with metal ions have been found to be successful in achieving biocidal activity [278,283].

The most established technique of modifying Chitosan has been to generate a positive charge by the addition of a quaternary ammonium group [278]. A broad range of investigations has determined that quaternised chitosan demonstrated much lower 'minimum inhibitory concentration' and higher resistance against *E. Coli, Staphylococcus aureus*. Likewise, in recent years the efficacy of carboxyalkyl chitosan has also proven to inhibit fungal biofilms namely *Candida giabrata, Candida krusei* etc. [283]. Several approaches have been used to prepare Chitosan doped with metals or metal nanocomposites with reduced agglomeration [286]. Chitosan can also act as both reducing and stabilizing agents that can prevent growth of *S. aureus* and *E. Coli* bacteria. Zhang *et al.*, [287] prepared Chitosan-TiO_2 composite with the idea of photocatalytic antimicrobial activity and found that the composites showed higher efficiency in inhibiting the growth of *E. Coli, S. aureus, A. niger* bacteria.

Another vital application of Chitosan is its usage in bone cement. Evidence shows that the use of chitosan in bone cement boosts the injectability and being cationic the electrostatic interaction prevents the growth of bacteria [288]. Pimentel *et al.*, [289] and Ustinova and Nikiforova [290] reported that Portland cement with Chitosan can be used to increase slurry handling time and with addition of nano-clay composites it can increase strength. Despite numerous advantages, Chitosan is not used in the construction industry which is commonly due to a lack of multidisciplinary knowledge across the field of engineering and there is a considerable scope to use chitosan in construction.

4.2.2.3. Cellulose doped with other antibacterial agents. Cellulose is the most abundant available biopolymer utilised in many applications such as packaging, textiles, sensors and soft actuators [291]. Cellulose and chitosan have similar molecular structures, with the same b-glycoside linkages. The main variation between them is the existence of primary amino groups at the C-2 positions in chitosan, where cellulose has hydroxyl groups [284]. Blended with cationic group (nisin), nanometallic (Ag-Cu) group, cellulose is found to have better biocidal activity especially against *E. Coli* and *fungus* [283]. It was also reported that cellulose doped with silver nanoparticles demonstrated high inhibition against *Proteus mirabilis, E. faecalic etc.* [292]. At present, there is a growing interest in utilising nanocellulose materials doped with metal and metal nanoparticles.

4.2.2.4. Polypeptides. Polypeptides such as keratin, collagen, caseinates etc. are attractive antimicrobial materials for food processing, packaging, textiles, dressing of burn wounds [283]. Despite its advantage of being biocompatible and biodegradable, its usage is limited. Among all polymers, polylactic acid (PLA) is currently used and considered as a promising one to replace petroleum derived polymers in making bioplastics. Unlike others, PLA is extremely compatible with blending of metallic nanomaterials and is being used in the production of thin films. A multilayer film fabricated using layer-by-layer assembly (LBL) with PLA peptides was found to be effective against *Micrococcus luteus* and more suitable for food preservation and coatings for implant devices [293]. Antimicrobial peptides (AMPs) are naturally occurring polypeptide sequences comprised of cationic and hydrophobic amino acids (~12–50 residues) with direct antibacterial activity [294]. Their application in the medical field has been present since their discovery in the late of 1980 s [115].

Construction application: Several antimicrobial compounds extracted from natural polymers are found to be effective against bacteria and viruses. Unlike chemical polymeric or nano-based materials, the natural compounds do not produce toxic effects [23]. Extracts from *Punica granatum, Vitex tri- folia, Chromolaena odorata, Aloe barbadensis,and Azadirachta indica* have biocidal properties [295]. Also, the combination of eugenol, eugenol acetate, carvacrol, thymol, and vanillin, and *Melaleuca alternifolia*, have been studied for their antiviral properties [295]. Despite the advantages, the exploitation of use of antimicrobial natural polymers (peptides, polysaccharides) can be majorly seen in medical, textile and food industries. Very scanty investigations have been carried out in other fields of application.

Interestingly, some researchers have made calcium-based cement using eggshells and reported that the cement's biocidal activity is effective and more specific for *Streptococcus mutants and E. faecalis* [245]. Little *et al.*, [52] reported that the addition of positive charge chitosan to any synthetic polymer will result in enhances corrosion resistance coating. Several researchers studied the use of natural and man-made polymers to coat various types of paper products [296,297] and found that they can damage bacterial cells within 3 h and in 9 h against microorganisms in the air and in food. Vartiainen *et al.*, [298] conducted investigations incorporating Chitosan with nisin and lactic acid coating on cellulose paper and antimicrobial activity was determined by inhibition zone method with agar diffusion assay against *B. subtilis*. It was discovered that compared to nisin, Chitosan with lactic acid inhibited the growth of *B. subtilis* instantly. Interestingly, there are a few researchers, who have proved that the use of natural essential oil extracts made from cloves, cinnamon and oregano also work well against *Penicillium nalgiovense, C. albicans, Aspergillus flavus, Eurotium repens, E. Coli and Bacillus cerus* [299]. Shankar and Rhim [296] prepared biopolymer coated paper using a ternary blend of alginate, carboxymethyl cellulose and carrageenan from grape seed and tested against pathogenic microbes such as *Listeria monocytognes* and *E. Coli*. Also, extracts obtained from grapefruit seed is more effective than others. Vishnuvarthanan *et al.*, [300] prepared paperboard from cow dung and banana fibres. The addition of hydrogen peroxide to the pulp enhanced the biocidal activity against *E. Coli* and *S. aureus*. Recently, many

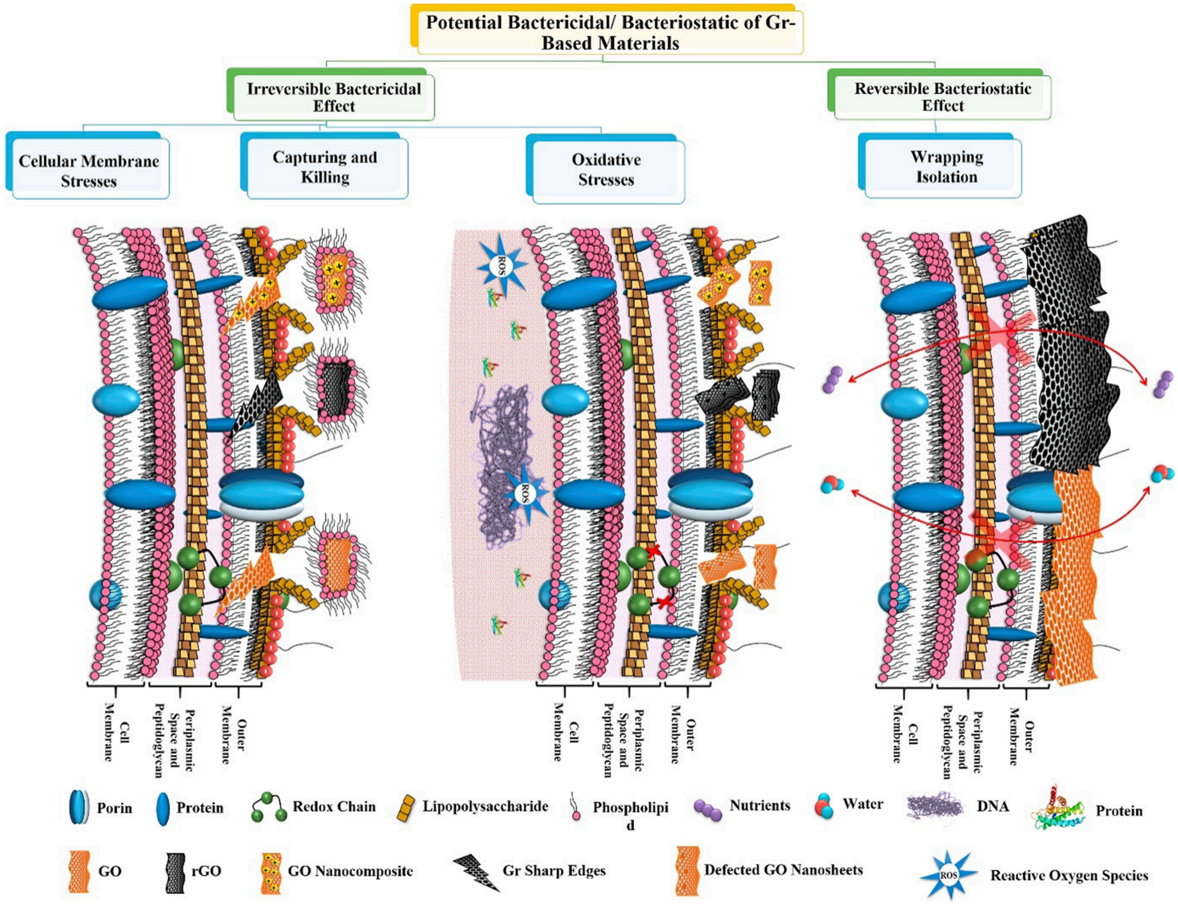


Fig. 18. Antimicrobial mechanism of graphene based materials [309].

researchers tested the effectiveness of biogenic corrosion inhibitors on mild steel and observed that it is an effective, environmentally friendly and sustainable treatment. One such biogenic corrosion inhibitor, *Tubinaria ornate* (seaweed) extracts were tested by Krishnan *et al.*, [301] in concentrated HCl medium on mild steel and they found 100 % mortality of bacteria in 12 h. Additionally, corrosion caused by fungal infestation *Cladosporium, Penicillum* and *Aspergillus spp*. can be protected using coatings embedded with nanosized metals or nanosized metal oxide particles, e.g., nanocopper oxide or nanozinc oxide [9,302]. In order to provide solutions to toxicity of the antimicrobial material and environment friendly material, *flavonoids, alkaloids, glucosides and tannins* are some green corrosion inhibitors (include amino acids, ionic liquids and plant extracts) that exhibits better antimicrobial activity against many gram-positive and gram-negative bacteria and are used as coatings for steel [303].

4.3. Other inorganic materials

There is a growing trend of using organic (carbon based) antimicrobial materials which include graphene, graphene oxide and multi-walled carbon nanotubes (MWCNT). These materials have a bactericidal effect on various kinds of species of bacteria [304].

4.3.1. Graphene oxide (GrO)

Graphene is a 2D material comprising of sp^2 carbons [305]. Owing to its exceptional physical properties such as a large specific surface area and mechanical strength, electrical properties, graphene has become a favourable nanomaterial [98,306]. Antibacterial properties of graphene have been explored since 2010 [307]. The antimicrobial activity of graphene involves both physical and chemical modes of action [308]. It is reported that there are three main mechanisms at play which adds to biocidal activity of Gr based materials and these are (i) membrane stress; (ii) oxidative stress, and (iii) wrapping isolation [309]. These mechanisms can act separately or together to inhibit bacterial growth. Fig. 18 shows a schematic illustration of antimicrobial mechanism of Gr based materials. Despite various advantages of graphene and its derivatives, researchers have found that graphene can cluster due to its very low water dispersion which makes it difficult to use [310]. Some researchers have also reported that doped polymers from graphene composites were able to kill microbes within an hour [308]. Gr-based coatings have also been shown to be effective in the marine industry as they can eliminate algae and corrosion in pipes [311]. Researchers have noticed that graphene-based coatings are one of

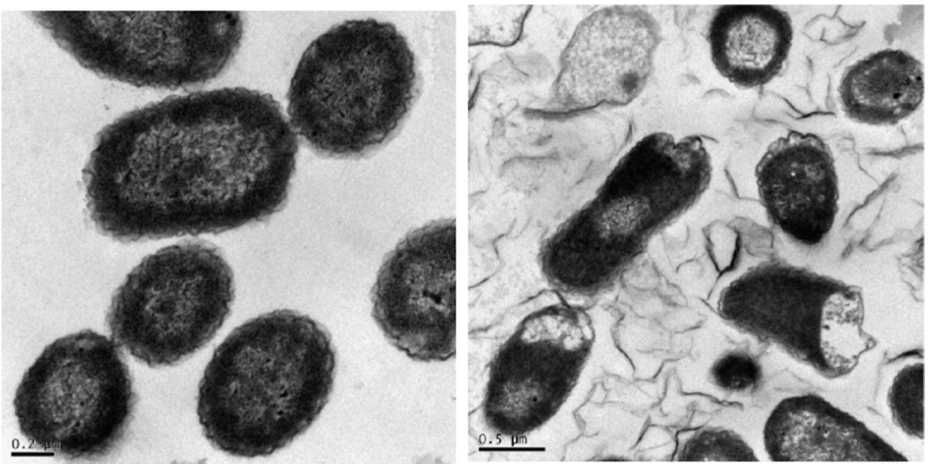


Fig. 19.. TEM images of rGO nanosheet papers before and after exposure to *E.Coli* [324].

the best solutions for microbiologically induced deterioration of concrete [50].

4.3.1.1. Carbon nanotubes (CNT). Carbon nanotubes (CNTs) are described as hollow cylinders formed by rolling a 2D graphene sheet [286]. In 2007, it was first discovered that single wall carbon nanotubes (SWCNTs) possess strong biocidal activity against *E. coli* [312]. Many researchers have shown that antibacterial activity of CNTs depends on their diameter, length, aggregation degree, concentration, surface functionalization, degree of purification, and time and intensity of contact [17,313]. Many researchers have also observed that SWCNTs exhibit strong antibacterial activity through direct physical interaction and aggregated SWCNT osmotic action. This leads to excessive oxidative stress due to residual metal ions in the CNTs, interfering with bacterial DNA replication and killing 80–90 % of bacteria within minutes [314]. The coatings containing carbon nanotubes exhibit uniformly low reflectance over a wide range of wavelengths from visible to far infrared [315]. The efficiency of CNTs can be improved by doping ZnO to enhance the photoactivation mechanism. With this mechanism, 100 % inactivation of bacteria was observed after 10 min of UV irradiation [312]. It was also observed that SWCNTs displayed the highest ROS generation than MWCNT, thereby killing a high percentage of bacteria in less time [316]. From the literature, the major antibacterial mechanisms of CNTs can be summarized as:

- Interruption of bacterial cell membrane by formidable electrostatic interactions among microbial exterior surface and CNTs, leading to oxidation of the membrane.
- ROS generation can destroy biological molecules of bacteria and/or indirectly trigger DNA damage.
- Impurity components (e.g., metallic NPs, catalysts, suspension) that are established into CNT-structures during production processes can contribute to their antibacterial activities.

4.3.1.2. Geopolymer concrete (GPC). Geopolymer is an inorganic material produced from activating materials rich in aluminosilicate such as fly ash, metakaolin, slag, etc., with strong alkaline solutions which induces geopolymerisation process [317]. GPC has become a promising green concrete due to its low carbon footprint and excellent mechanical properties [57]. GPC reduces the consumption of natural resources, possess cost-effectiveness, and has capacity to form different structural configurations and to remain intact for extended periods without repair works [318]. Some of the well-known examples where GPC has been widely explored are pavements, roads, bricks and sleepers [319]. Recently, GPC was also found to be a potential multi-functional material for wastewater treatment. GPC possess combined positive properties of vitreous ceramic pipes (acid, permeability resistance) and improved mechanical properties which makes its suitable against MICC [57]. GPC exhibit high acid resistance and provides stable barrier against microbial deterioration. Unlike conventional OPC and Ca-rich binders, GPC avoids the formation of Ca-rich acid dissolvable hydration products allowing it to have high acid resistance [59]. While the chemical resistance of OPC based materials is governed by acid buffer capacity of their constituents, GPC relies on an ion exchange reaction [26].

Many researchers consider GPC as analogous to zeolite due to its similarity in nanostructure but packed in an amorphous microstructure that enables embodiment of antimicrobial cations to reduce bacterial colonisation [58]. It is also demonstrated by many researchers that doping of metallic or nanometallic ions to GPC results in a substantial improvement in its bactericidal activity. Hashimoto *et al.*, [58] and Drugă *et al.*, [12 12]postulated that after acid attack, the metal ion (Cu^{2+}) can release gradually as per cation exchange reaction and they can interact with microbes resulting in DNA damage of the bacteria. Another study revealed that the use of nano silver-silica modified GP mortar causes ROS production and cell wall rupture of microbes which explains the plausible mechanism in its antimicrobial activity [187]. A few studies have also alluded to the benefit of GPC as being a sustainable solution for crack maintenance i.e., incorporating bacteria to achieve self-healing properties [320,321]. Khan, *et al.*, [322] compared occurrence of MICC due to low Ca-fly ash-based GPC and sulphate resistance OPC. They observed that widespread gypsum crystallisation and precipitation led to an increase of MIC for the sulphate resistance OPC than GPC.

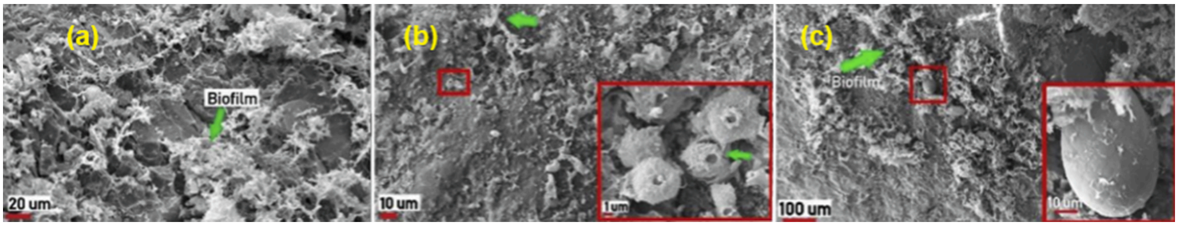


Fig. 20. SEM images showing the formation of biofilms on (a) OPC; (b) CAC; (c) GP mortar exposed to wastewater [12].

Table 2
Antibacterial solutions used to mitigate MIC in sewer concrete pipes.

Antibacterial agents	Effects	References
Bio-concrete (calcite precipitation)	Bacterial cells incorporated precipitates $CaCO_3$ which self-heals the cracks formed due to MIC in sewer concrete pipes	[330]
Nitride	Formation of free nitrate acid (FNA) in calcium nitrate concrete shows promising way to mitigate MIC	[329]
Magnesium hydroxide ($Mg(OH)_2$) based coating	$Mg(OH)_2$ neutralises the H_2SO_4 acid produced during MIC thus inhibiting corrosion	[244]
Geopolymer concrete	High resistance to acidic environment aids in inhibiting MIC in concrete pipes	[59,332]
Nitrate reducing bacteria (NRB)	NRB limits H_2S production and increases redox potential, but it is suitable only against SOB not SRB	[52]
Nano copper oxide	Higher leaching rate of Cu ions suppress the growth of *thiooxidants*	[333]
Nano zinc oxide	Photocatalytic and hydrophobic nature of nano ZnO embedded in concrete mitigates *thiooxidants* and *thiobacillus*	[204]
Zwitterion polymers	Superhydrophilic nature found to prevent direct contact between bacteria and surface and thus repel the SRB	[271]
Aluminium ions	Addition of calcium aluminate cements in sewer concrete pipes are found to be effective in inhibition of MIC	[334]
Graphene	Oxidation and acid resistance of graphene make it suitable for inhibition of MIC in sewer networks	[325]
Silver embedded zeolites	Sewer concrete containing Ag-zeolites found to be effective in killing *Acid-thiobacillus* and *thiooxidants*	[254]
Sodium tungsten	Complete inhibition of growth of SRB and SOB with just 50 μM nutrient solution	[335]
Nickel ions	Incorporation of Ni in concrete binds to plasma membrane and inhibits sulfurdioxygenase and oxidation of *thiooxidants*	[336]

Although significant research efforts have already been made to establish the antimicrobial activity of GPC, these studies so far have discussed only empirical GPC tests on acid resistance. There is no record of studies comprising the actual performance of GPC related to MIC and resulting bio-chemical degradation from a microbiological perspective.

Construction application: Graphene has been used as a support to disperse and alleviate various nanomaterials, such as metals, metal oxides, and polymers, with high antibacterial efficiency due to the synergistic effect [323]. Graphene based nanocomposites have wide range of applications in medical, food, water disinfection, industries and marine applications. Hu *et al.*, [324] reported that advantages of production graphene oxide and reduced graphene (rGO) based nanosheet papers is that they inhibit the growth of *E. coli* and fungal growth. *E. coli* grown on paper were killed by destroying their membrane integrity, which was then confirmed by transmission electron microscopy (TEM) studies [324] as shown in Fig. 19.

Some authors have determined that the dispersion of graphene as filler particles into the coating matrix to form a graphene composite anti-corrosive coating enhances the performance and life of the materials [76,311]. Therefore, graphene based composite coatings are a promising strategy to prevent corrosion in metals. One such example can be identified by the study carried out by Krishnamurthy, *et al.*, [325] wherein polymer infused with rGO based coating is highly resistant to MICC. Hassan *et al.*, [326] reported that incorporation of CNT fibers in reinforced concrete aid in bridging of cracks formed due to corrosion and thus increases the bonding of steel to concrete. Additionally, CNT fibers rupture the microbial cell adhered on the surface of the reinforced concrete. Likewise, Chuah, *et al.*, [327] pointed out that the addition of CNT fibers into concrete increases the functionality and mechanical properties of concrete.

Hashimoto *et al.*, [58] and Grengg *et al.*, [59] observed that the geopolymer concrete is a green sustainable approach towards MICC in sewer pipes due to their high resistance towards acidic environments. Drugă *et al.*, [12] compared the growth of bacteria strains on ordinary Portland cement (OPC), Calcium aluminate cement (CAC) and Geopolymer (GP) cement matrix against MICC when exposed to wastewater for 35 days. GP develop less biofilm on their surface, followed by CAC, and worst is OPC as shown in Fig. 20. Both OPC and CAC showed clusters of bacteria forming biofilm whereas in GP, bacteria was invertebrate eggs/resting stages (Fig. 20-c).

Gutiérrez, Ruby Mejía de *et al.*, [328] investigated antibacterial activity of geopolymer based cocktail mortar. It comprises of glass waste, titanium oxide (TiO2) and 5 wt% copper oxide (CuO) NPs. They observed that the cocktail mixture inhibits the growth of *Staphylococcus aureus, Escherichia coli, and Pseudomonas aeruginosa* within 24 h. These results indicate the possibility of generating antibacterial surfaces by applying geopolymer composite especially in the region susceptible to biocorrosion. Similarly, several researchers observed that addition of different metallic NPs in geopolymer concrete mixture can result in self-healing and sustainable material against microbial colonisation [187,317,320].

Table 3
Summary of different manufacturing methods used to deposit antimicrobial materials.

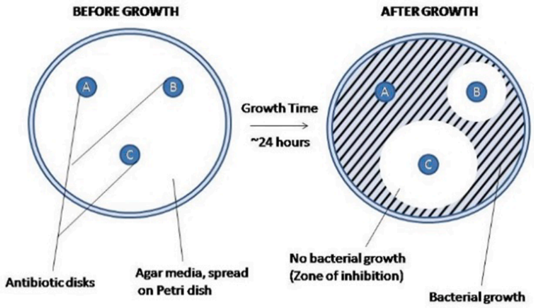
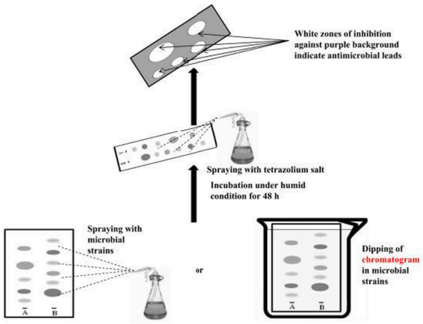
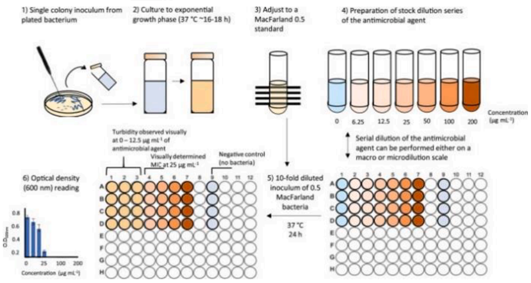
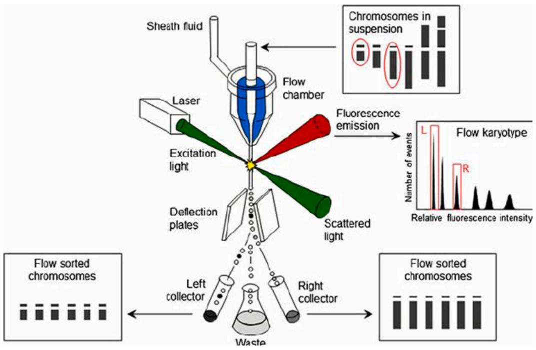
Method	Description	References
Mechanical methods (Brush, Pad, and Roll Coating)	It is a manual or automatically controlled method to apply antimicrobial materials especially in liquid or gel form to surfaces using brushes, pads, and rolls. Generally, the coating is formed by air drying. However, extra care and repeated maintenance is required for this type of coating.	[91,342]
Dip coating	Dip coating implies immersing a material into a tank comprising coating agents, and then allowing it to drain. It is dried using heat, air, or UV radiation. One of biggest drawback of this method is not being able to control the coating thickness.	[343,344]
Flow coating	Ideal for complex geometries and involves application of antimicrobial material into the substrate via freely flowing media. As the coating flow freely over the surface, the entire surface gets coated consistently. Unlike dip coating, very little coating material is required as any excess recirculates within the system.	[91]
Spin Coating	The substrate is usually rotated in this method to spread the coating material by centrifugal force. It is widely used for microfabrication of functional oxides on glass surfaces, or crystals.	[91]
Electroplating	Metals and poly-ionic polymers are applied by electroplating to conductive materials, such as aluminium and steel. The substrate is submerged into a metal salt or a poly-ion solution in an electrically conductive tank. It is then cathodised for deposition of metals and positively charged polymers and anodized for negatively charged polymers. The film thickness can be monitored by the reaction time and the concentration of the solutions.	[345]
Electroless plating	In this method, the substrate is put into the metal salt solution and a reducing agent, like hydrazine. The elemental metal is then deposited onto almost every material surface, which does not react with the metal salt solution in water.	[346,347]
Acid-etching	The process of cutting a hard surface, such as metal, utilising an etchant. Although this method is commonly evaluated, a major problem with this method is its tendency to deposit residual ions on those metallic surfaces that can leach easily.	[342]
Plasma spraying	Plasma spray is a thermal spray coating method utilised to produce a high-quality coating using a combination of high temperature and high energy heat source, with a comparatively inert spraying in argon, and high particle velocities. Usage of this technology allows spraying of almost any metallic or ceramic on to a large range of materials with good bond strength, while diminishing deformation of the substrate.	[118,174,348]
Cold spray	Powder particles (typically 10 to 40 μm) are accelerated to very high velocities (200 to 1200 $m.s^{-1}$) by a supersonic compressed gas jet at temperatures below their melting point to bombard on the substrate. The particles undergo rapid plastic deformation which disrupts the thin surface oxide films that are present on the surface of metals and alloys. Thus, it creates strong mechanical interlocking between the substrate and antimicrobial materials.	[349]
Physical vapour deposition	Physical vapour deposition (PVD) is a process used to produce a metal vapour that can be deposited on materials as a thin, pure metal or alloy coating. The process is carried out in a vacuum chamber at medium to high vacuum (10–6 torr) using a cathodic arc source.	[342,348]
Chemical vapour deposition	Metals, and their oxides are deposited onto surfaces by chemical modification of a gaseous organic compound into solid particles. The transition of the gaseous compound can be achieved by thermal decomposition, pyrolysis, reduction, oxidation or hydrolysis process.	[350]
Sol-gel deposition	Sol-gel method requires the substrate to be dipped into a colloidal solution (sol), which gradually undergoes polycondensation, leading to the formation of a thin gel layer on the surface. This method is low cost and entails minimal processing temperatures, which aids to retain the bulk mechanical properties of the material.	[351,352]
Laser texturing or coating	An emerging technique that has been used to prevent bacterial adhesion. There are basically-two ways by which the killing of microbes are carried out.(i) It is used to etch nanoscale grooves into the metal, increasing its surface area and enhancing its bactericidal properties.(ii) It is used for depositing the cationic charged material that weakens the bacterial membranes and produces high mechanical stress leading to physical disruption of the outer membrane.	[353,354]

Another promising method studied by Li *et al.*, [329] to attenuate the MIC in concrete sewers using nitrate admixture concrete and established a negative relationship between the nitrite admixture concrete and the abundance of SRB using DNA sequencing and increased pH on the surface. Ground breaking research studies were carried out by Chetty *et al.*, [330] wherein bio-concrete incorporating self-healing bacteria (calcite precipitate bacteria) where used to control the cracks formed due to biocorrosion. In a alkali activated concrete made out of fly ash, silica fumes and micro silica inhibits the microbial growth owing to its refined pores and stable gel structure [331].

Some of the possible antibacterial solutions to attenuate MICC in concrete sewer pipes, observed by various researchers are given in the Table 2.

For the past few years, many researchers have used radiopacifying agents such as bismuth oxide (BO), zirconium oxide (ZO), bismuth carbonate (BC), tungsten, etc. as antimicrobial agents, impregnated in white Portland cement [337]. Many desirable characteristics of these radio pacifiers were established especially in medical fields for extending the opacity of the cement in implantation or retrograde root canal filling. in the place of mineral trioxide aggregate (MTA). Weckwerth *et al.*, [338] evaluated antimicrobial activity through interference of the radio pacifiers BO, BC, and ZO in white Portland cement by radial diffusion method against *S. aureus, C. albicans, Aeruginosa and E. Faecalis*. It was noted that the radio pacifiers showed bactericidal effect only against *C. albicans* by creating a zone of inhibition not for any other microbes. Conversely other researchers have inferred that addition of ZO or BO or calcium tungsten displayed bactericidal effect by producing zone of inhibition against all microbial strains [339,340]. It was also

Table 4
In-vitro assessment of antimicrobial materials [355].

Test methods	Description
1. **Disk Diffusion Methods** a. Agar diffusion method b. Antimicrobial gradient method (Etest) c. Agar well diffusion method d. Agar plugs method e. Cross streak method f. Poisoned food method	
2. **Thin Layer Chromatography (TLC)-Bioautography** a. Direct bioautography b. Agar overlay bioassay	
3. **Dilution Methods** a. Broth dilution method b. Agar dilution method	
4. **Time-Kill Test**	It is the most used method for determining the microbial impact on materials. It is a strong tool for obtaining information about the dynamic interaction between the antimicrobial materials and the microbial strain. The time-kill test reveals a time-dependent versus microbiological induced concentration (MICC)
5. **ATP Bioluminescence Assay**	It is based on the ability to quantify adenosine triphosphate (ATP) produced by microbes. Light is emitted during the reaction with the luciferin-luciferase reaction, which is captured using biosensors. Amount captured is detected as presence of growth of bacteria.
6. **Flow-cytofluorometric Method**	

inferred that powerful bactericidal action was achieved by inhibiting cell growth and respiration even at low concentration. Despite various advantages of addition of any radiopacifying agents in cements, most esearchers have found they reduced physical properties like setting time, porosity and pH [337]. Therefore, it has been suggested that addition of two or more radio pacifiers or silver NPs will enhance both biological and physical properties of Portland cement [185,341].

4.4. Methods of application of antimicrobials

There are various types of manufacturing technologies available by which these antimicrobials can be deposited to the substrate. Not all these methods are readily adapted in construction (especially on-site construction) but detailing these methods are provided to give a glimpse on the prospects that each method in Table 3.

5. Performance evaluation and influential factors

Building materials consisting of various types of antimicrobials have been used and introspectively studied under various environmental conditions related to different types of microbial contamination. I*n-vitro* evaluation of the efficacy of antimicrobials has been performed and efforts have been made to establish a better understanding of their biological activity.

5.1. Performance evaluation

Many assessments, such as disk-dispersion, well-dispersion and broth or agar dilution are some of the well-known and routinely used methods but time-killing testing, flow cytofluorometric and bioluminescent techniques are also other popular methods. Table 4 describes the different types of *in-vitro* laboratory-based evaluations that are paid attention to in the literature.

5.2. Factors affecting antimicrobial activity

The activity of antimicrobial materials against microorganisms depends on several factors, some of which are intrinsic qualities of the materials, others of which are the environmental and microbial factors. Recognition of these factors should lead to better use of antimicrobial materials. Some of the important factors are briefly reviewed below:

Microbial factors: Antimicrobial activity of different materials depends on the type of microbes. Different microbes have different sensitivity to antimicrobial materials. For example, gram negative bacteria with a thinner cell wall can be killed more rapidly with less concentration of agent than that of gram-positive bacteria with a thicker cell wall [7]. The choice of antimicrobial material also depends on the growth of microbial adhesion. Many researchers have observed that out of four phases of bacterial growth (lag, log, stationery and death) [356] it is important to kill/inhibit bacteria in the preliminary stage itself, otherwise the time lapse to kill them will increase resulting in multiple growth of bacteria. Yet another important parameter for choosing antimicrobials is population size or microbial density. Larger population of microbes requires longer time to kill, in that due course of time, bacteria can mutate and develop antimicrobial resistance (AMR). Therefore, it is necessary to develop a antimicrobial substrate that can kill rapidly and also inhibit further proliferation of bacteria [357].

Intrinsic factors: There are many intrinsic factors such as concentration, wettability, surface topography, molecular weight, contact time and type of monomer attached which affect the suitability and mechanism of bactericidal action of different materials. It is well reported that higher concentration of antimicrobial materials can have greater efficiency in killing microbes. In higher concentrations, antimicrobial materials produce ROS causing oxidative stress on bacterial cells, that causes significant damage to the cell membrane, degrade important proteins and nucleic acids and initiate lethal stress response to bacteria [76]. Wettability is another important parameter to determine the efficacy of antimicrobial materials. Mostly, the lower wettability of materials with higher hydrophobicity (low surface energy) may impose better bactericidal properties than hydrophilic surfaces. In contrast, a few studies have illustrated that excessive hydrophilic substances decreases cell adhesion and spreading by bacterial repulsion [96,358]. Many researchers have tried to manifest the relationship between surface roughness and bacterial adhesion. Most of the studies have proposed that increase in surface roughness increases hydrophobicity of the materials, thus resulting in reduction of bacterial adhesion [359] and vice versa for hydrophilic materials. Researchers have noticed that application of antimicrobial materials using laser, plasma, thermal spray increases the surface roughness of the material that can kill the bacteria rapidly [358,360]. The chemical composition of antimicrobial surfaces can also change bacterial adhesion. In one of the study, it was reported that crystalline antimicrobial surface are more effective in killing microbes than amorphous surface [361]. Materials like polymers (Chitosan) performances depend on its molecular weight and density. Higher the molecular weight, higher the bactericidal activity of materials [284]. In case of polyzwitterion materials, the type of attached to the polymer network, bond strength, surface charge density decides the nature of mechanism of bactericidal activity. Biocidal activity of photolytic monomer activates only in the presence of UV rays and cationic monomer such as QAC are found to be more effective in zwitterion polymers [362]. Many studies have also noted that the longer the exposure time, the more the biocidal activity of the materials. Also, prolong contact time prevents the further adhesion of bacteria.

Environmental factors: Temperature, pH, light, humidity etc. are some of the critical parameters that affect the mechanism of antimicrobial materials. Higher temperature usually increases the effectiveness of the antimicrobial materials. Conversely, a few studies have identified that the bactericidal activity of the Ag NPs before heat treatment were more active than heat treated silver nanoparticles [363,364]. Many researchers have explored smart antimicrobial materials that are triggered with an increase in temperature and release in low temperature [265], found to be very effective in hot and humid regions.

pH is another important parameter that antimicrobial materials are dependant on. Most of the studies have determined that, at neutral pH, antimicrobial materials are cationic and kill bacteria by disrupting the phospholipid membrane [7,365]. However, more acidic or alkaline medium have a negative effect on inhibition of bacterial growth. It is also very well-known that acidic medium (pH < 7) causes corrosion in concrete and steel, wherein studies have highlighted that the growth of SRB is more susceptible when pH is < 7.

Table 5
Commercially available antimicrobial solutions used in the construction industry.

Name of the Product	Manufacturers	Active Substances	Remarks
Silvershield ZP Tech Excalibur	Microban International ltd., USA [372]	Triclosan, Silver ion, Zinc ion, Copper ions and QAC	Coatings carried out on different types of building products such as ceramic titles, floorings to remove mildew and fungus
Agion	Sciessent LLC, USA [373]	Copper + Silver ions	Leveraging Cu and Ag (zeolite) biocidal activity, designed to release during bacterial growth and used on walls. Ceilings and rood linings
Interpon Biocote	AkzoNobel, Netherlands [374]	QAC impregnated with epoxy or zeolite	Powder coating done on walls, toilets, furniture, etc. and protect against bacteria and moldews
Touchclean	Dortrend International ltd., United Kingdom[375]	Titanium oxide	Nanotechnology antibacterial coatings applied on walls, doors, floors etc. and works effective against MRSA, SARS and E. coli.
Biomaster	Addmaster, United Kingdom [376]	Silver ions	Paints and powder coatings for walls, floors and ceilings
SteriTouch	Steri Touch, ltd., United Kingdom [377]	Silver ions	Additives and coatings designed to reduce growth of mould, fungi, E. coli and Salmonella and used mainly on laboratories, tables, hospitals etc.
Nippon	Nippon paint and coatings, Osaka, Japan [378]	Cuprous ions encased in special glass ceramic powder	Innovative coating solutions for architectural decorative panels, marine structures, hospitals and effective against S. aureus and viruses
Dow Du Point	Dow Chemicals, USA [379]	Polyurethane, Silver and silver oxide ions	Provide safety in HVAC, removes mold and algae growth on roof lining, wall panels
PPG coatings	PPG, Industries, Inc., USA [380]	Ag ions + onium compound (halogen ions with Br ions) + carboxylic acid resin	Provides electrocoating, powder coating, liquid paint for industries and heavy loaded floorings and effective against S. aureus, pseudomonas aeruginosa and non-enveloped viruses
Sanitised PL 29–36	Sanitsed, AG, Switzerland [381]	PVC, Silver	Antimicrobial protection against yeast, fungus, mildew, algae growth for floorings, swimming pools and roof liners
Dialogforoum Nano	BASF, SE, Germany [382]	Zinc, silver salts	Nanomaterial paintings and varnishes used in church redecorations, industrial flooring, wooden floorings and walls
Hygentic and Ultrason	BASF SE, California [382]	Styrene butadiene copolymer with silver ions	Paintings and coatings for hospitals medical devices and water tanks

A few studies have reported differently. For example, Saliani, *et al.,* [366] found higher antibacterial activity of ZnO NPs at acidic pH levels with the maximum toxicity at pH = 4 and pH = 5 for *E. Coli* and *S. aureus* at 42°C.

Light is an important parameter for a photocatalytic material. Generally, in the presence of light (sun light or UV) ROS species are produced that destroys DNA and regenerative cells of the bacteria. In one of the studies, five different wavelengths (each with three intensities) in the visible spectrum against gram-positive and gram-negative biofilms were evaluated and observed that violet and blue light showed better bactericidal effect than green, yellow and red light [367]. Light responsive materials usually, perform the antimicrobial activity in the presence of UV light (wave length < 400 nm), which accounts for only a small proportion of solar light (3–5 %) [211]. It has been determined that modified antimicrobial substances having lesser band gap will involve less energy UV radiation and this is more sustainable approach [368].

Many researchers have observed that decay rate of bacteria increases with increase in humid condition. However, no significant responses were noted in lower humidity conditions [369,370]. Also, dry and humid condition are better for airborne bacteria, wet and low humid for aqueous inhibited bacteria (plankton) [371].

6. Remarks and future direction

Industries worldwide have started to promote the development of antimicrobial solutions. Some of the key industries trying to develop antimicrobial construction solution from across the globe identified by this review are shown in Table 5. Most of these were found to be based in the UK and the USA. These companies produce different varieties of solutions in the form of coatings and additives that prevalently destroy *fungus, algae, mildews, E. Coli and S. aureus* microorganisms. Microbial deterioration has been a great threat to any construction material (or) structures. The relationship between antimicrobial parameters such as content, retention rate and dispersion are still required to be thoroughly investigated. Combination of two or more antimicrobial agents to perform a synergistic biocidal activity can be used as a wise strategy. The toxicity due to leaching of biocidal agents into the environment needs to be tackled so there is a strong need to develop eco-friendly and biosafe antimicrobial building solutions. A grey area of concern is the antimicrobial resistance (AMR) which needs urgent attention, particular around the construction sector. In addition, it is more beneficial to create smart building materials that have at least two destruction mechanisms and eradicate all kinds of microorganisms. Currently, most studies are limited to experimental investigations and more modelling efforts are required. The use of artificial intelligence and digital tools can help to steer better in this direction.

As such microbial degradation has been a great threat to all industries, particularly the construction industry. This threat can be so serious that it can be damaging to the economic stability, human health and the economy. The review highlights those antibacterial materials can damage cell membranes either by producing reactive oxygen species (ROS), physical contact (electrostatic interaction), or the release of metal ions, which interferes with the DNA and causes cell death. Metallic nanoparticles such as TiO_2, ZnO works under the principle of the photocatalytic mechanism which can further be boosted by adding zwitterionic polymers. Also, silver embedded zeolites were found to possess high biocidal activity against different microorganisms. Therefore, many researchers considered it appropriate to use them to protect concrete in sewer pipes from biocorrosion. The use of chitosan seems to hold more promise in future construction activities and is an area that needs immediate research. We also need to assert whether industrial antimicrobial additive mixing, or in-situ site mixing of antimicrobial mixing is a safer practice. Intuitively, as-supplied industrial material with a deterministic antimicrobial performance would seem to be a better option, but research in this topic is yet to be done.

Despite numerous advantages, there are some limitations of having antimicrobial solutions, the foremost of which is the risk of inhalation, environmental pollutions etc. This alludes to the need of safe handling of antimicrobial materials at site especially in harsh environments. In addition, with recent advances in technology, the use of 'smart' antimicrobials that can kill two or more germs at a time should be used in all applications. Also, the addition of a natural sustainable polymer will make the current construction practice more sustainable, biosafe and human friendly. Overall, this review revisits most of the studies reported on this topic. It draws attention to the possible antimicrobial mechanisms, discusses the key challenges and provides future recommendations for developing new, efficient antimicrobial materials that can provide sustainability and safe construction.

Data statement

As this is a review paper, no new data was generated.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

As this is a review paper, no new data was generated

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