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Recent design approaches, adhesion mechanisms, and applications of antibacterial surfaces

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ABSTRACT

Adhesion of bacteria on surfaces and the resultant biofilm formation is a great risk to human health care services, food packaging and storage, and the marine industry. This has necessitated the need for the scientific community and industries to continually develop ways of fabricating surfaces that can prevent bacterial adhesion, kill adhered bacteria, and prevent/disrupt biofilm formation. Therefore, this review is focused on providing an up-to-date discussion on the recent strategies adopted by researchers in the design of antibacterial surfaces. It highlighted the different surface characterization techniques, discussed the mechanism of bacteria adhesion and biofilm formation, reviewed the recent method of fabricating antibacterial surfaces and their industrial applications, emphasized the advantages and drawbacks of these techniques, and proposed outlooks for future designs.

Introduction

Nowadays, biomaterial surfaces with efficient antibacterial properties are in crucial demand. This is because the adhesion of bacteria on surfaces commonly leads to the formation of bacteria colonies resulting in the evolution of biofilms [1]. Bacterial biofilm is a great risk to human healthcare services and industrial operations which include medical equipment and surgical devices, the public health environment, food

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packaging and storage, water filtration systems, and so on [2,3]. The actual report suggests that about 80% of human bacterial infections are related to biofilm. This is because bacteria in the biofilm community are not often detected during diagnostic procedures, they are reported to have increased resistance against antibiotic remedies and can protect the bacteria from the host immune defense system [4,5].

It is interesting to note that surfaces like bed rails, door handles, touch plates, phones, computer keyboards, etc., which are common hospital and office items are prone to touch and can be highly fouled by microbes. For instance, it has been shown that germs such as *Escherichia coli (E. coli)*, and *Staphylococcus aureus (S. aureus)* can stay on the aforementioned items for months [6]. Although constant and efficient cleaning, together with good hand hygiene, reduces bacterial transmission. Nevertheless, complete eradication remains a big challenge. In addition, with the global escalation of antibiotic-resistant microorganisms like methicillin-resistant *S. aureus* (MRSA) strains, serious nosocomial infections have become a major concern for hospitals or other allied centers. The United States data showed that approximately 720, 000 hospital-acquired infections (HAI) were recorded in 2006, leading to $125 billion in added hospital bills. These observations highlight the need for innovative methods of managing sanitation in hospitals and allied centers, hence antimicrobial surfaces are potential strategies for complementing present-day hygiene measures [7,8].

Furthermore, amid the growing demand for medical implants and devices such as catheters, contact lenses, and cardiovascular and prosthetic devices, one major drawback is the high danger of bacterial infection because it is a common and serious complication in the hospital that often results in death [9]. It must be emphasized that the immune system of any patient receiving a medical implant is already compromised and most times the surgical operations that are done to insert implants leave surgical wounds that create an environment prone to bacterial infection. Besides, the potential source of bacteria is the medical implant and the surgical devices, therefore, blocking the adherence of bacteria on the implants and the devices is the first line of deterrence in preventing bacterial infection.

*Listeria monocytogenes (*L. *monocytogenes), E. coli,* and *S. aureus* are among the common contaminants found on food contact surfaces (FCS) [10]. These microbial contaminations can result in severe complications in the food processing and packaging industries. Reports have shown that the build-up of food, protein, and minerals on the FCS can increase the susceptibility of biofilm formation which has a detrimental impact on the quality of food, plant performance, and efficiency [11]. In 2019, the World Health Organization (WHO) reported that more than half a billion persons suffered ill health, and about 420,000 die yearly as a result of contaminated food [12]. For that reason, the adoption of antibacterial materials in the design of FCS has been described as an effective strategy to prevent bacteria contaminants [10,13]. In the marine industry, bacteria are the most significant microbes on marine surfaces, and because they are early colonizers, their cells influence the framework and function of the well-developed biofilm [14,15]. Besides, bacteria, diatoms, and other microorganisms are responsible for microfouling; these microbial pollutions on marine surfaces grant an easy platform for other marine species (algae, barnacles, and mussels) to attach and breed resulting in macrofouling [16]. Biofouling (micro/-macro fouling) has a huge environmental impact and significantly increases the cost of operating and maintaining marine-related industries such as maritime transport, desalination plants, petroleum industries, aquaculture, etc. [17]

Since the property of a surface (topography, roughness, wettability, charge, etc.) plays an important role in how the surface interacts with bacterial [18,19], it becomes necessary to continuously devise means of disrupting such interactions in order to significantly minimize the adhesion of bacteria on surfaces and prevent the formation of biofilms. It is good to note that significant research has been carried out in this regard. Herein, we explored the current design techniques as illustrated in Fig. 1 which researchers have adopted. Firstly, a list of different analytical techniques used in bacteria and biofilm characterization was presented in Table 1, followed by a discussion on the working mechanics of bacteria adhesion and biofilm formations. Thereafter, the different design strategies and their applications in the medical, marine industry, and food processing and packaging were reviewed and emphasis was made on the advantages and drawbacks of the different techniques while proffering suggestions for future designs.

Mechanism of bacterial adhesion and biofilm formation

The mechanisms of biofilm formation are reported as a multifactorial and complex process. The main process in biofilm formation consists of adhesion/adherence/attachment, aggregation/accumulation/maturation, and dispersal/detachment phases as illustrated in Fig. 2 [42–44]. Several pieces of literature on the mechanism of bacterial fastening and the formation of biofilm have shown the adverse effect and advantages of this formation on metals [45–47].

According to Lee et al. [48], it was detailed that micro-organisms have a distinctive capacity to adhere to the surface of some embedded medical devices, forming biofilms that negatively affect device efficiency and further escalate the danger of multiple drug-resistant issues. The physicochemical characteristics of most biomaterials have over some time been acknowledged to perform an essential task in the formation of biofilm. It was noted in the review that a series of findings in

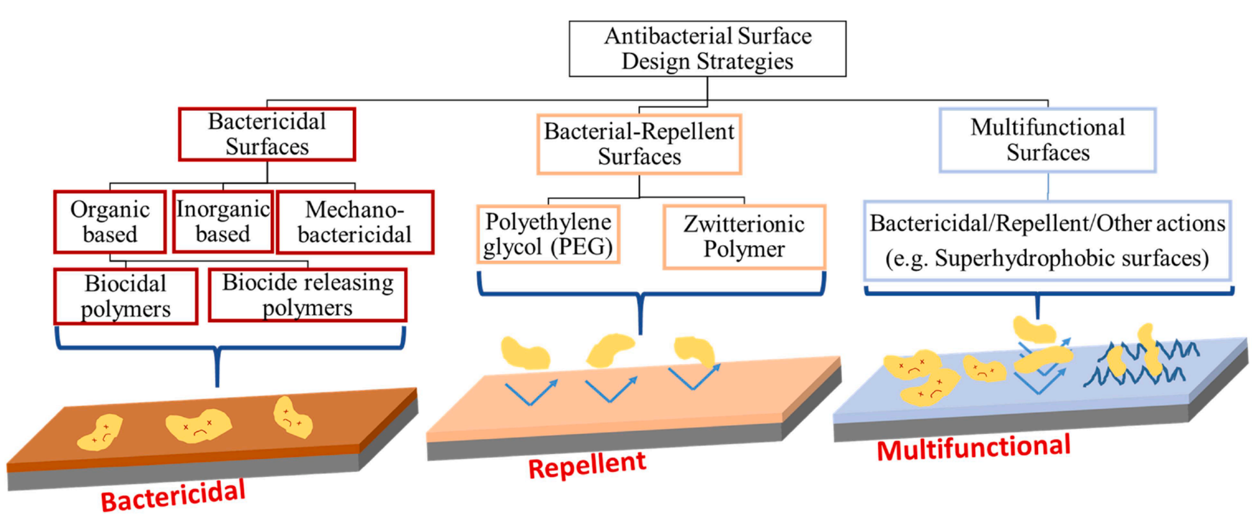

Fig. 1. Illustration of antibacterial surface design strategies.

Table 1
Different analytical techniques for assessing bacteria and biofilm surfaces.

S/N	Technique	Application	Reference
1.	Staining assays	The use of dyes to stain the cells or backgrounds to add color and contrast for easy visualization.	[20–22]
2.	Genetic assays	Enables the efficient identification of the existence of definite genetic sequences linked to different bacteria species. Example; Polymerase chain reaction (qRT-PCR)	[21,22]
3.	Fluorescence-based methods.	Employ the UV range electromagnetic spectrum to study single cells (examples; flow cytometry and fluorescence microscopy), biofilm not in suspension (example; full-field imaging), biofilm in suspension (example; multiplate reader fluorimeter and dip probe)	[23,24]
4.	Raman Spectroscopy	The Raman spectrum offers bacterial-specific molecular fingerprints which can be used for analysis.	[25,26]
5.	Optical Microscopy	Rapid and non-destructive visualization and characterization of cells with minimal sample preparation.	[27,28]
6.	Confocal Laser Scanning Microscopy (CLSM)	Enables the scanning of thick biological samples; Characterizes the structural features of the biofilms, gives the 3D configuration of the biofilm, and identifies live/dead cells.	[22,29]
7.	Scanning Electron Microscopy (SEM)	Visualization of cells and biofilm at high magnification	[30,31]
8.	Focused Ion Beam (FIB)	Used in combination with SEM. FIB enables the cutting of target sections of the sample, getting rid of the exposed surface areas, and/or milling the cross-sections to study the inner of the biofilm.	[32,33]
9.	Transmission Electron Microscopy (TEM)	Visualization of bacteria surface architecture and size at nanoscale resolution	[31,34,35]
10.	Energy-Dispersive X-ray Spectroscopy (EDX)	To obtain local compositional spectra and plots of cells and biofilms	[36,37]
11.	X-ray microscopy	Enables high-resolution imaging and compositional mapping of cells and biofilms without any detailed preparation and with decreased radiation compared to electron microscopy.	[38,39]
12.	Atomic Force Microscopy	High-resolution visualization of bacteria cells and biofilm surfaces, measurement of adhesive force and elasticity of the biofilms, and observation of bacteria multiplication processes and production of extracellular polymeric substance (EPS).	[40,41]

the global world has significantly aroused great ideas in the adoption of 3-dimensional surface topography to set up antibiofouling designs that defy bacterial colonization. Developing evidence has shown that the surface topography of most medical devices, especially those modeled for tissue integration, can unintentionally enhance microbial attachment. Regardless of the number of literature on biofilm control and surface topographies, there is still an uncounted connection between how numerous bacteria perceive and react to the 3D surface contour and the intelligent design of antibiofouling materials. Impelled by this gap, the authors concluded that 3D surface topography, either unintended or intended, is one of the important factors to consider in the rational model of safe and environmentally friendly medical devices [48]. The graphical representation of the review is shown in Fig. 2(B).

In related research done by Feng et al. [49], the authors presented work on expediting the *Pseudomonas aeruginosa* biofilm formation through an exogenous N-Acy-L -homoserine lactones (AHLs) stimulation: Focused on the regulation of the bacterial motility, metabolic activity, and adhesive behavior. Recent reports have shown that quorum sensing (QS) is of broad biogeochemical significance, it can affect microbial metabolism by controlling cell-to-cell interactions and coordinating group behaviors via the process of production, release, and population-wide detection of autoinducer signaling molecules such as AHLs. The research elaborated on the AHL-supported quorum sensing (QS) system and showed that it exhibited unique ecological importance in the development of bacterial biofilm. The research examined the aspect of exogenous AHLs on the *Pseudomonas aeruginosa* biofilm development. Fig. 2 (C, D) shows variations in biofilm-related metabolites at different N-hexanoyl-homoserine lactone (C6-HSL) and N-octanoyl-homoserine lactone (C8-HSL) concentrations for extracellular polymeric substance (EPS), Rhamnolipid, Pyocyanin and the possible mechanism for how exogenous AHLs help *P. aeruginosa* develop biofilms more quickly. The results obtained showed that both the C6-HSL and C8-HSL enhanced the formation of *P. aeruginosa* biofilm up to about 2.47 and 1.88 times, respectively. Additional investigation proved that exogenous AHLs assisted immensely with the adhesion capability rather than the growth rate. Furthermore, the metabolic activities and bacterial motility were uniquely upgraded by AHLs. Also, the functional genes of the microbes (i.e., rhlR, rhlI, lasR, and lasI,) that participated in controlling the development of the biofilm were greatly shown in AHLs reactors. These results gave an extensive understanding of the functions of AHLs in providing insightful guidance, and mediated biofilm formation on the management of biofilm in most wastewater treatments through the biofilm technology [49].

Significant research done by Etim et al. [50] showed the mechanism of adhesion of bacteria and the formation of biofilm on reinforced steel by sulfate-reducing bacteria (SRB) in a mild alkaline simulated concrete pore solution. The findings showed that SRB adhered to the surface of the steel after 28 days of immersion. With the deployment of a bacteriostatic agent known as organic silicon quaternary ammonium salt (OSA), the bacteria adhesion was mitigated. The corrosion mechanism showed a large pit depth of about 36.70 μm in the medium containing SRB but reduced to 3.31 μm in the medium containing SRB and OSA. The study proves that OSA as an effective bacteriostatic agent is capable of mitigating the excessive adhesion of SRB on the steel and thus limits the corrosion rate as shown in Figs. 3 and 4 [50].

According to Liu et al. [51], it was established that bacterial assembly was possible at the early phase of adhesion in wastewater treatment of biofilms (Fig. 5). The results showed that the start-up of biofilm was very essential and basically a slow process, especially in moving bed biofilm reactors (MBBRs), with the exact operation starting with the micro-organisms constantly bound on surfaces. Although much research addressed the critical process focusing majorly on building models established on single strains, the proposal of the unsteady adhesion mechanism of structured microbial communities is yet to be fully investigated. In the study, the unsteady adhesion actions of structured bacterial communities gotten from the aerobic tanks of about eight comprehensive wastewater treatment plants (WWTPs) was evaluated using impedance-based real-time cell analysis (RTCA). The results showed the time range of the unstable adhesion to be from ~8.85 to ~17.06 h, showing meaningful disparities in bacterial colonization characteristics. Adopting the principal components analysis (PCA), K^+, $Na^{+,}$ and proteins were shown to largely affect the biofilm unstable adhesion processes. Comprehensively, the unstable adhesion found in MBBRs was controlled by the prevalent bacterial breed and the easement of environmental parameters by repulsive forces, supplying potential actions of dosing QS signals and decisive cations at the early adhesion stage in the reactors, to promote the initial formation of biofilm.

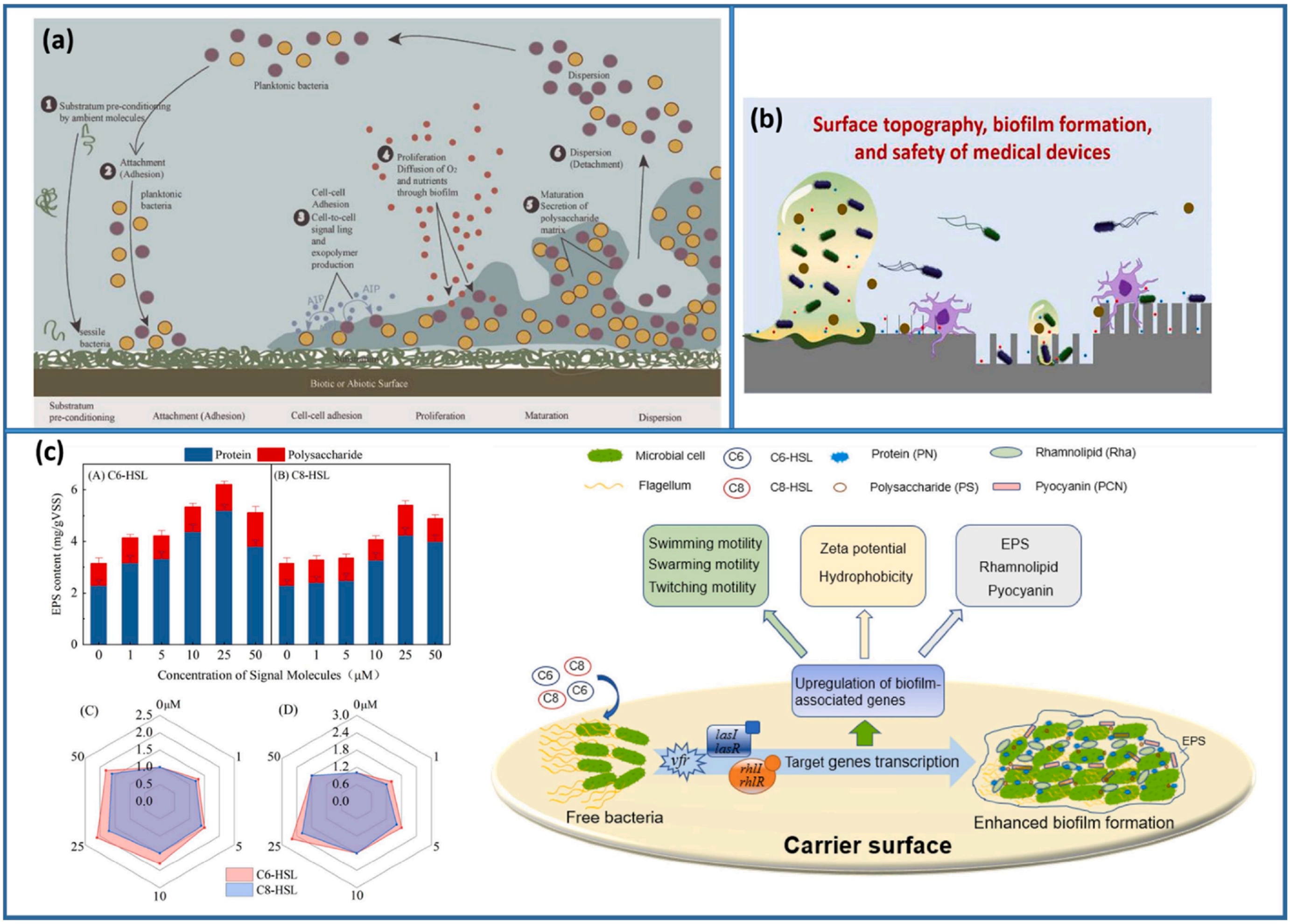

Fig. 2. (A) Different levels of biofilm formation; start with the adhesion of bacteria to a surface that serves as a substrate for microbial attachment, then continues with aggregates of cell-cell adhesion [42]. (B) A schematic representation of how surface topography affects biofilm formation. There is significant biofilm formation on the section of the substrate without a pattern [48]. Changes in biofilm-related metabolites at various C6-HSL and C8-HSL concentrations. (A) and (B) EPS; (C) Rhamnolipid; (D) Pyocyanin, and (E) possible mechanism [49].

Factors affecting bacteria adhesion

Bacteria are first transported towards a surface followed by adhesion and subsequent formation of biofilms. However, the transportation of bacteria toward a surface is dependent on the nature of the environment and the type of bacteria [52]. Motile bacteria can easily move to the surface and glide sketchy paths close to the surface in order to enhance adhesion, whereas immotile bacteria depend on the gravitational force, hydrodynamic forces, and Brownian motion to reach a surface [52,53]. When bacteria reach a surface, adhesion to the surface can be categorized into a two-stage process: The first stage is the initial reversible attachment while the second stage is an irreversible attachment [54]. The thermodynamic method of evaluating microbial adhesive interactions normally considers the interfacial free energies between the interacting surfaces. It suggests that spontaneous adhesion of the microbial cell to the substrate surface happens when the free energy per unit area (ΔG_{adh}) is negative (i.e. $\Delta G_{adh} < 0$), while adhesion is energetically unfavorable when the free energy per unit area is positive (i.e. $\Delta G_{adh} > 0$) [55,56]. Theories on the adhesion of colloidal particles like the Derjaguin–Landau–Verwey–Overbeek (DLVO) and extended DLVO (xDLVO) model have also been employed to interpret bacteria adhesion [57,58]. The DLVO theory takes into account the attractive Liftshitz–van der Waals (LW) and the repulsive electrostatic double layer (EL) interactions [59]. These interactions change with the separation distance between the substrate's surface and the bacteria. When the bacteria attain the separation distance of secondary minimum energy, reversible adhesion tends to occur. Overcoming the energy barrier, the bacteria can get closer to the substrate and achieve irreversible adhesion [60]. DLVO theory usually fails to predict the interaction between a bacteria and surface at very small separation gaps, where Lewis acid-base interactions are prevalent [59]. Lewis acid-base interactions are made up of hydrophobic and hydration effects. Therefore, the xDLVO theory accounts for the Lewis acid-base interactions together with DLVO forces for characterizing total interaction between cell and surface. It requires stronger bacteria-surface interactions to move from reversible to irreversible adhesion. These stronger interactions usually depend on bacteria surface appendages like lipopolysaccharide (LPS), EPS, and fimbriae [52]. To design an effective antibacterial surface, it is necessary to understand the relation between bacteria adhesion and surface properties (such as surface roughness, surface wettability, and surface charge).

Surface roughness

Surface roughness is essential to bacteria attachment to surfaces prior to biofilm formation. The presence of nanoscale roughness decreases the physicochemical potential barrier a bacterial cell encounters when it reaches the surface [61]. Also, nano roughness can decrease bacteria adhesion forces by decreasing the contact area between the bacterial cell and the substratum surface [62,63]. Lu and co-workers demonstrated the influence of surface roughness on the adhesion of

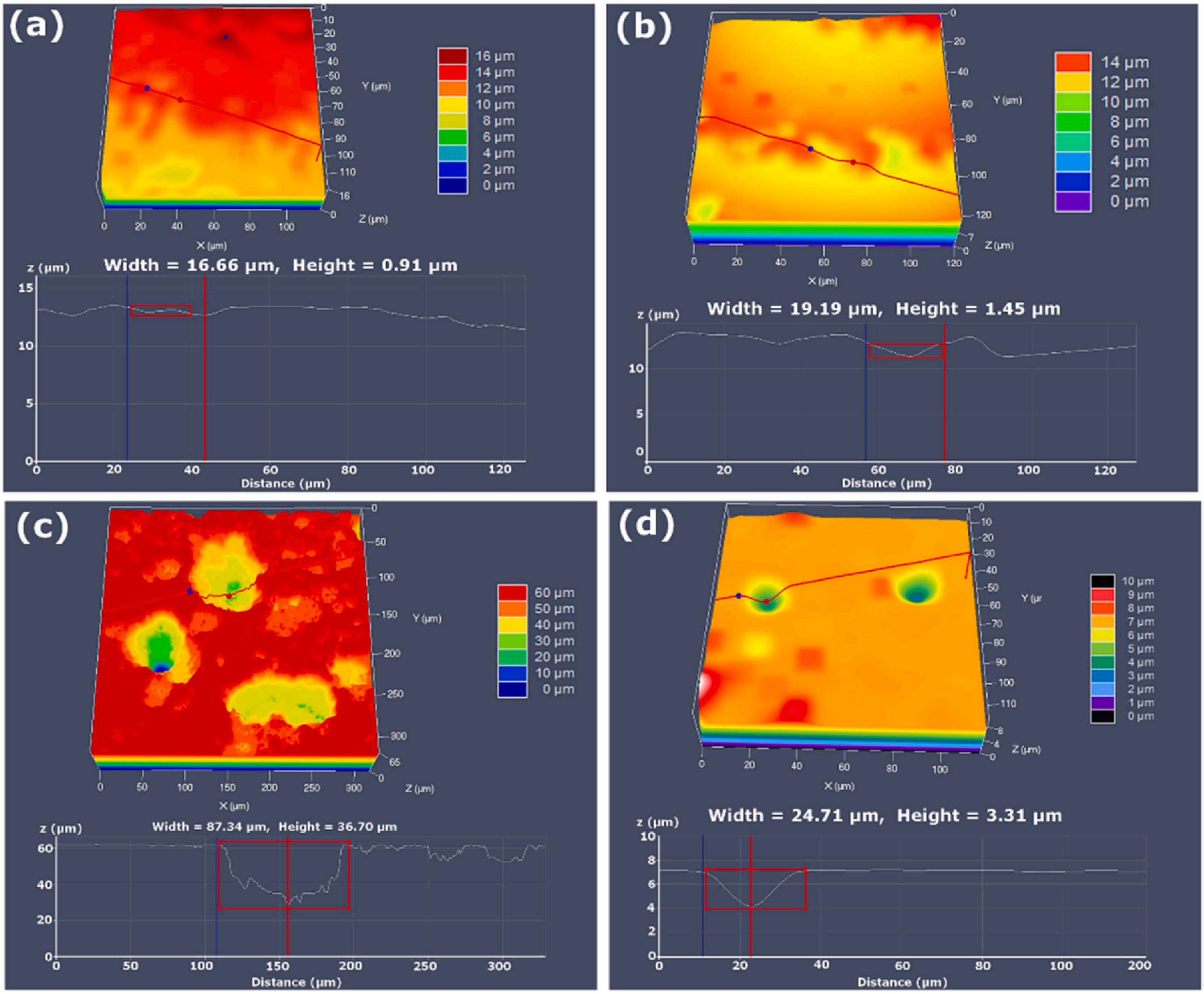

Fig. 3. A pictorial representation of the pith differences due to SRB adhesion on the reinforced steel. (a) sterile (STR) medium, (b) sterile with antibacteriostatic agent, (c) SRB medium, and (d) SRB with antibacteriostatic agent. In Figures (a) and (b) the pit formation varied slightly while large pit was formed in Figure (c) compared to other media because of the presence of SRB. However, it's seen in Figure D that the pit formation was reduced when antibacteriostatic agent was added [50].

S. aureus on yttria-stabilized zirconia bio-ceramic surfaces [64]. They confirmed that the number of bacterial adhesion on a surface with average roughness (R_a) of 1 nm roughness is just 2% of that on a normal surface with R_a 205 nm . As the surface roughness moves from the submicron scale to the nanoscale level, the surface wettability was shown to increase making it difficult for the adhesion of hydrophobic *S. aureus*. Also, the anchoring points for bacterial adhesion either decreased or disappeared hence weakening the bacterial-surface bonding strength. Spengler et al. found that the adhesion of *S. aureus* decreased as the size of the nanostructures increased [65]. They explained that the macromolecules of the bacteria cell can only tether the top area of the surface, hence, increasing the nanostructure reduces the area available for tethering.

Although, it is suggested that bacteria adhesion has a negative correlation at nanoscale level roughness, however, different range of nanoscale roughness shows unique bacteria adhesion behavior according to the study done by Yang et al. [52]. For instance, under static conditions, there is preferential adhesion of bacteria on surfaces with R_a 0.23 – 6.13 nm range which might relate to the cellular metabolic activities like improved EPS production on smoother surfaces and the size of bacteria [52]. For relatively higher roughness ($>$ 6 nm), bacteria were shown to prefer adhering to rougher surfaces [66]. The larger roughness tends to provide shelter for attached bacteria to withstand the fluid shear stress [67]. In addition, bacteria-surface interactions have been found to be dependent on bacteria species; *P. aeruginosa* cannot attach to a smooth surface with Ra $>$1 nm [68] whereas the adhesion of *S. epidermidis* was not affected by the roughness [69].

Contrary to the adhesion behavior at nanoscale roughness, studies have shown a positive correlation with the submicron or micron scale roughness [52,70]. Santhosh et al. employed different micromachining finishing techniques to prove the decrease in the adhesion of *E. coli* and *S. aureus* on stainless steel and titanium alloy as the surface roughness decreases [71]. Interestingly, Wassmann et al. [72] found that the R_a values have no effect on the adhesion of *S. epidermidis*, neither on titanium nor on zirconia surfaces. They suggested that it is inadequate to evaluate the effect of surface morphology on initial bacterial adhesion using only the R_a value. It must be accompanied by an additional 3D analysis of the microstructure. Their suggestion supports the works done

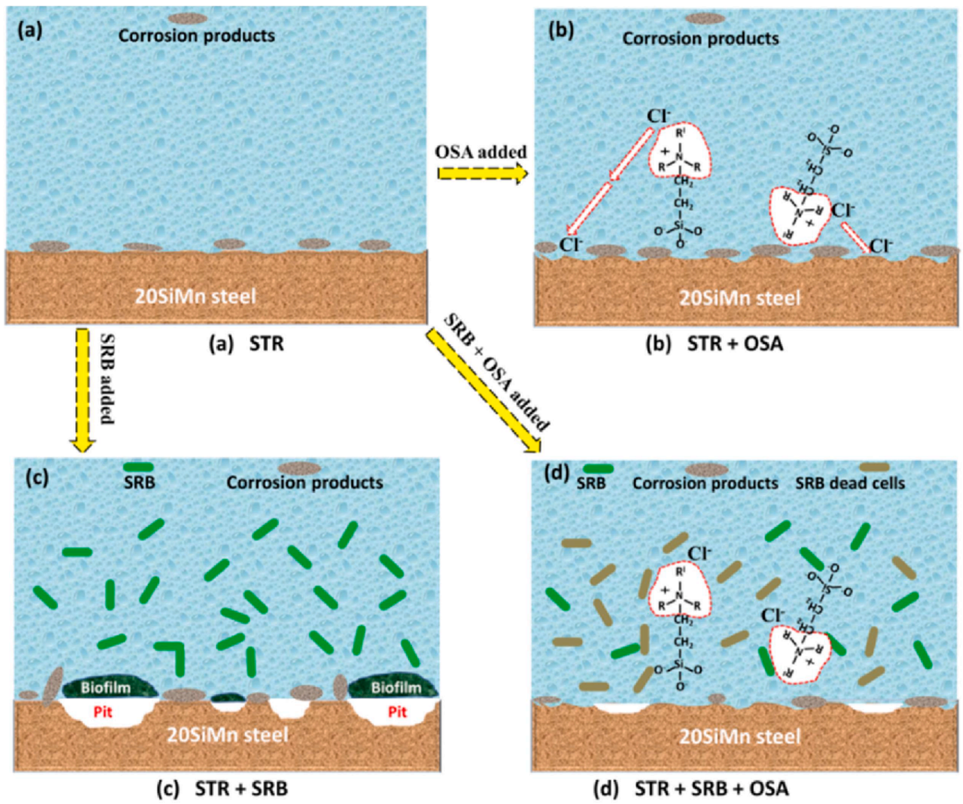

Fig. 4. A schematic representation of the mechanism of SRB adhesion on the reinforced steel surface. The corrosion mechanism depicts the antibacteriostatic agent feature in the study media. The ammonia component was able to mitigate the SRB growth while the other component adhered to the surface of the substrate [50].

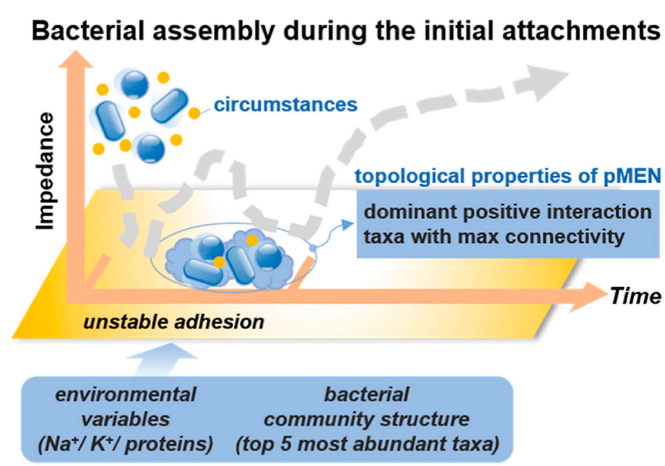

Fig. 5. The mechanism of the bacterial assembly during the early attachment process [51].

by Barbour et al. [73] and Taylor et al. [74]. In addition, Katsikogianni and Missirlis proved that the adhesion of bacteria does not only rely on the sizes of surface asperity but also their shapes, and bacteria prefer to adhere to surface topography features as large as their own diameters [75]. An average bacteria cell such as the rod-shaped *E. coli* is about 2 μm long and 0.5 μm in diameter, *S. aureus* is about 1 μm in diameter, the smallest bacteria (e.g. *Mycoplasma pneumonia*) ranges from 0.1 to 0.25 μm in width, and the largest bacteria such as Azobacter is 2 – 5 μm in diameter [76]. So, surface features (scratches, corrugations, and roughness) that are of the same magnitude as the size of the bacterial improve the contact area and hence the binding potential, while surface features that are larger/wider than the bacterial size reach the binding potential of a flat surface. Features that are too small for the bacteria to fit (< 100 nm) reduce the contact area of the bacterium and thereafter, the binding potential [75]. In summary, nanoscale roughness has the best antibacterial adhesion whereas submicron and micron-scale roughness facilitates bacterial adhesion. Flat/smooth surfaces are known to majorly rely on other material properties [52].

Surface wettability

The wettability of a surface has been reported to play a major role in bacterial adhesion. Feng et al. developed a medical indwelling catheter with specific wettability (inner hydrophobic/ outer hydrophobic etc.) exhibiting good antibacterial properties [77]. Kong et al. prepared a wettable polycaprolactone-chitosan/chitosan oligosaccharide (PCL-CS/COS) nanofibrous membrane for wound dressing application. Their results showed efficient antibacterial properties attributed to the high hydrophobicity (~130°) of the PCL layer [78]. Increasing hydrophobicity has been shown to increase antibacterial efficiency [79]. Since bacteria may need EPS to attach to a surface and develop biofilms, it is expected that higher surface hydrophobicity will suppress initial bacteria attachment since the larger portion (~90%) of EPS is water and the remaining parts include nucleic acids, protein, polysaccharides, and various ions. Notwithstanding, some reports have suggested that hydrophobic surfaces can decrease the bacteria near-wall velocity via collisions thereby enhancing settling and subsequent adhesion [52,80]. Hydrophobic interplay is a factor in both stages of bacteria adhesion and a major influence in enabling the initial bacteria adhesion [81]. The most common case is that more hydrophobic cells attach more strongly to hydrophobic surfaces, whereas more hydrophilic cells attach more strongly to hydrophilic surfaces [82,83].

Furthermore, superhydrophobic surfaces are known to possess trapped air in the nanostructures which could reduce the bacteria adhesion force and hinder surface adhesion [84]. The air layer can also decrease the contact area between the surface and the bacteria cell thereby hindering adhesion [85]. Peptidoglycan (PGN) and lipopolysaccharide (LPS) are part of the essential components of the cell wall in Gram-positive and Gram-negative bacteria, respectively, where they act as the major regulators of bacteria attachment. The resistance of superhydrophobic surfaces to PGN and LPS is due to their unique hierarchical structures where air packets are trapped in nanoscale interstices leading to a minimized hydrophobic interaction between the bacteria and substrate [86,87]. Mu et al. have done significant work in analyzing the bacterial reaction/deposition kinetics in both hydrophobic and superhydrophobic regimes [88]. They also identified three key factors necessary for antibacterial properties on superhydrophobic surfaces. Firstly, the presence of air packets indicates the existence of a Laplace pressure force acting toward the bacteria, so the Laplace pressure force should surpass the bacterial adhesive force. Secondly, the diminished effective substrate area prevents direct contact with the bacterial cell due to the air packets. Thirdly, the decrease in the van der Waals attractive force that keeps the adhering bacteria on the substrate.

On the other hand, literature has shown that superhydrophilic surfaces possess a low degree of bacteria attachment as well [89,90]. The strongly bound water layer developed on the superhydrophilic surfaces hinders the interaction between the surface and bacteria cell preventing the formation of biofilm [86]. In comparison to superhydrophilic and superhydrophobic surfaces, surfaces with moderate wettability adsorbed more LPS leading to greater bacterial attachment as reported by Jiang et al. [91].

Surface charge

Bacterial cells are known to exhibit negative charge in liquid environments due to the presence of phosphate, carboxyl, and saccharide groups on the surface of the cell, hence negatively charged surfaces can repel bacteria through the electrostatic repulsion effect. However, recent work done by Huan et al. proved the efficient antibacterial property of highly positive arginine carbon dots [92]. The positive charges on the surface of the carbon dots enabled electrostatic interaction with negatively charged bacteria, disrupting the normal physiological activities of the bacteria by altering the bacteria membrane integrity and the leakage of small molecular compounds inside bacteria resulting in bacterial death [92]. One major technique for measuring the surface charge of particles or solids is the Zeta potential, also known as the electrokinetic potential [92,93]. The surface charge also depends on the pH value and ionic strength of the solution [94,95]. Researchers are currently using polyelectrolytes to modify the charge of solid surfaces thereby improving the antibacterial effects [96–98].

Different strategies for designing antibacterial surfaces

This section discusses the different approaches used by researchers to design antibacterial surfaces.

Bactericidal surfaces

Antibacterial surface coatings that have a bactericidal effect can kill bacteria before or after contacting the surface [2,9]. Based on their chemical composition, bactericidal surfaces can be divided into two categories: organic and inorganic. Organic active surfaces are principally made of polymers and can be grouped into two depending on their action mechanisms: biocidal polymers and biocide-releasing polymers [99]. The inorganic surfaces consist of metal oxides and nanoparticle-based metals as well as non-metals such as graphene, carbon nanotube, and selenium [9].

Organic based bactericidal surfaces

Biocidal polymers. Biocidal polymers exist as a form of contact-based antimicrobial system, where antibacterial agents are coated on the surfaces to kill fastening bacteria. Biocidal polymers typically contain either a quaternary ammonium group or a cationic functional moiety, both of which make the polymer chain a whole antiseptic [100]. Quaternary ammonium silane of 3-(trimethoxysilyl)-propyldimethyloctadecyl ammonium chloride (QAS), quaternized poly(2-(dimethylamino ethyl) methacrylate) (PDMAEMA), and quaternized poly(4-vinyl-N-alkyl pyridinium bromide) (PVP) are three common biocidal polymers having quaternary ammonium groups in their side chains as shown in Fig. 6 [101–103]. According to the antibacterial mechanism of these polymers, the long chain cationic moiety first disrupts the outer lipopolysaccharide network of negatively charged microbial cells before moving inside to cause more cell damage [104].

Beyth et al. incorporated an antimicrobial based quaternary ammonium poly-ethylenimine (PEI) nanoparticles in dental resin and investigated the effect of the composite on the growth and the integrity of the membrane of the *Streptoccocus mutans*. The findings showed that quaternary ammonium PEI nanoparticles were immobilized in resin-based materials, creating a hybrid dental composite resin with excellent mechanical properties and strong antibacterial capabilities [105].

Kurt et al. fabricated contact-active coatings with surface-modified polyurethane (PU) polymers by thermodynamically driven surface enrichment of bactericidal species. In these studies, the composition and structure of naturally existing antimicrobial proteins varied, leading to an optimized formulation of alkyammonium PU coatings. These coatings are made of a PU backbone, PEG-like methoxyethoxyethoxy and trifluoroethoxy side chains resembling chaperone, soft block, and microbial-resistant surface-concentrating (alkylam) molecules [106]. Longer alkyl chains (C12), when compared to shorter chains, exhibited a higher biocidal effect (C6). Trifluoroethoxy side chains benefit from the effect of higher surface enrichment on the alkylammonium C-6 chain more than PEG-like methoxyethoxyethoxy side chains do. In addition, the authors uncovered that modifying structural and compositional properties, such as side chain length, charge group, and chaperoning are vital for the rational design of efficient contact-active PU coatings that can kill *S. aureus, P. aeruginosa,* and *E. coli* on contact [106].

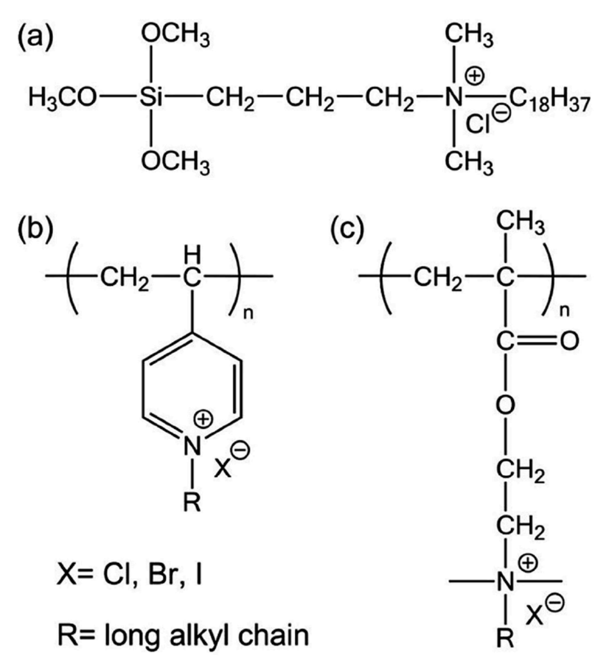

Fig. 6. Chemical structures of (a) 3-(trimethoxysilyl)-propyldimethyloctadecyl ammonium chloride (QAS), (b) quaternized poly(4-vinyl-N-alkylpyridinium bromide) (PVP), and (c) quaternized poly (2-(dimethylamino ethyl) methacrylate)(PDMAEMA) [9].

Biocide-releasing polymers. In biocide-releasing polymeric systems, antibiotics are directly embedded on the polymer's surface and serve as a carrier for biocides/antibiotics with an effective controlled release approach to halt bacterial activity. Natural and artificial polymers, such as poly (lactic-co-glycolic acid) (PLGA), poly (methyl methacrylate) (PMMA), polyethylene glycol (PEG), and dextrans, have been loaded with a variety of antibacterial agents, including antibiotic drugs (e.g. amoxicillin, norfloxacin, cephalothin, gentamicin, tetracycline hydrochloride), or germicides such as chlorhexidine and triclosan [107–109]. Active agents can be integrated into polymers physically through adsorption or layer-by-layer assembly, chemically through covalent bonding, or by polymerizing an antibiotic-containing molecule [110].

Chuang et al. have employed a layer-by-layer deposition approach to design polyelectrolyte multilayer films containing gentamicin (GS), an effective antibiotic against biofilms of numerous staphylococci [111]. The layer-by-layer fabrication approach incorporates charged small molecular species into the films without modifying the physical or chemical structures, rendering the film's construction simple and efficient.

In their study, the heterostructure composed of a biocompatible polyanionic hyaluronic acid (HA), poly (β -amino ester) (Poly X) capable of hydrolytic degradation, and GS was used, resulting in films with a tetralayer architecture $(((\text{Poly X/HA})_1(\text{GS/HA})_1)_n)$, where n represents the tetralayers' number. The electrostatic and hydrophilic interactions between the components of the film were modified by using Poly Xs of various molecular weights. Increases in the hydrophilicity and charge density of Poly X were accompanied by an increase in GS loading density, which may have been caused by greater electrostatic interactions within the films. Furthermore, these interactions controlled the rate of release of the antibiotic from the film, with a slower rate of GS release being caused by a stronger binding affinity [112].

Inorganic based bactericidal surfaces

Another means of impacting antimicrobial properties on a surface is by incorporating inorganic nanoparticles that either generate antimicrobial agents or kill the bacteria on contact. Several nanoparticles such siliver (Ag), magnesium oxide (MgO), titanium oxide (TiO_2), silicon dioxide (SiO_2), zinc oxide (ZnO), copper oxide (CuO), and platinum nanoparticles have been reported to possess good biocidal activity [2, 113,114].

Copper-endowed surfaces. Copper is known to exhibit antimicrobial properties [115,116]. Although not completely characterized, the cause of cell death due to copper ions is thought to be multifactorial. Copper is a catalyst for the production of reactive oxygen species (ROS) which can cause oxidative damage to cellular components. Free copper ions may alter protein conformation and damage cytoplasmic enzymes. Disintegration of DNA and depolarisation of the cytoplasmic membrane may also be major factors [117,118]. Thus, it has been proven severally that the efficacy of copper surfaces increases with the increased concentration of Cu in such surfaces. It is in this vein perhaps that the US Environmental Protection Agency has registered copper as the first solid antimicrobial material [119]. Although many hospitals and other public environments have incorporated copper surfaces within their design (e.g., copper handrails, door handles, toilet seats), there is no documented verifiable evidence to suggest this has reduced infection rates tremendously. However, in a study of single rooms, six high-contact surfaces originally manufactured from stainless steel, aluminum or plastic were replaced with copper alloy equivalents. The findings indicate that in comparison to patients in control (non-copper materials) rooms, patients occupying a room containing all six copper components for the duration of their stay had about 69% lower risk of acquiring infection [120]. This, therefore, points to the infection-reducing ability of Cu ions.

Silver-doped surfaces. Silver (Ag) containing materials, particularly silver nanoparticles (Ag NPs) have received huge interest in recent years because of their intrinsic expansive antibacterial spectrum and durable antibacterial activity on many surfaces [121]. Lee *et al.* have developed silver nanoparticle-integrated textiles and report their antimicrobial effectiveness in bandages used for wound healing as well as medical settings that are prone to bacterial attack [122]. Nunzia et al. recently confirmed ~100% antibacterial efficiency of filter masks prepared with Ag- and Cu-based coatings [123]. Even though the explanation of the antibacterial mechanism of Ag NPs still appears rather unclear, a perspective has been conventionally adopted suggesting that it is the silver ions (Ag^+) discharged from Ag NPs rather than the nanoparticle itself that performs a critical role [124]. It has been illustrated that the Ag^+ could also boost the permeability of the membrane and form reactive oxygen species (ROS), similar in behavior to the Cu ions, disrupting the bacterial cell walls and the resultant bacteria death. As the pioneer discoverer, Morones et al. [125] recently found that Ag^+ and antibiotic mix could encourage a synergistic effect which leads to decreasing or eliminating the drug-resistant bacterial strains, thereby broadening the antibacterial spectrum of the actual antibiotic drugs. Meanwhile, the consensus about the bactericidal ability of Ag implies that binding on the surface and causing damage to the cell membrane are key functions of bactericidal Ag. This is to some degree buttressed by the reports of the metabolic effects of Ag (I), in that it hinders various oxidative enzymes, metabolite efflux, yeast alcohol dehydrogenase, the intake of succinate by membrane vesicles, and the transport chain of *E. coli* and other predominant microbes on surfaces. The earlier researchers on AgSu were specifically interested in its macromolecular synthesis. They discovered that protein, DNA, and RNA syntheses were interrupted in AgSu-treated bacteria but were subdued in the cell-free systems [126]. In addition, Ag^+ is said to be more highly toxic to bacteria than copper ions, and consequently, their use as silver surfaces to limit bacterial spread within a hospital environment may be less appropriate owing to other associated health issues they may further exude.

Antibacterial-zinc surfaces. Zn nanoparticles have demonstrated strong biocidal activity against the pair of Gram positive and Gram-negative bacteria [127,128]. Perelshtein et al. used ultrasound process to deposit ZnO and CuO nanoparticles on cotton fabric surfaces, producing nanomaterial-based surfaces that are extremely effective against both *E. coli* and *S. aureus.* The antibacterial activity of ZnO nanoparticles may be attributed to the release of Zn^{2+} and its interaction with the interior of bacterial cells, which may damage or obstruct the transport mechanism across the cell membrane [129,130]. Zinc ion has multiple inhibitory effects on the intact bacterial cells' activities such as glycolysis, glucosyltransferase production and polysaccharide synthesis, transmembrane proton translocation, and acid tolerance. It is able to augment the ability of proton to permeate the membranes of the bacteria cells, decrease the synthesis of Adenosine Triphosphate (ATP) in glycolyzing cells, and reduce the activities of F-Type Adenosine Triphosphate Synthases (F-ATPase) owing to its potential to suppress the glycolytic enzymes glyceraldehyde-3-phosphate dehydrogenases and pyruvate kinase including the phosphoenolpyruvate [131]. Zinc oxide nanoparticles (ZnO-NPs) are bio-safe and not harmful to human cells. Also, zinc in the form of a nanoparticle is more lethal to bacteria than in micron forms, hence, the antimicrobial actions of ZnO-NPs can be a result of the likely interplay between the bacteria and the nanoparticles. Bacteria growth can be suppressed by ZnO-NPs action on the bacteria surface or inside cell penetration. This can lead to disrupting bacteria enzyme systems, dislodging magnesium ions needed for the bacteria enzymatic actions, and followed by an obvious bactericidal outcome. Zinc is used nowadays as coatings on household wares and surfaces because of both its bactericidal and galvanizing properties. It is commonly found in saliva and plaque and is effectively used in dermatological products like lotions, creams, and ointments [132]. In addition, zinc is added to oral health and dental mixtures such as mouth rinse and toothpaste, where it can stay for longer hours after usage when the concentration is much without attracting public health concerns [133].

Antibacterial carbon nanotubes. More recently, carbon nanotubes (CNTs) have attracted significant attention in biomedicine and have been employed as promising antimicrobial agents due to the combination of their remarkable mechanical properties and exceptional antibacterial activities [134]. The bactericidal potential of these CNTs is dramatically influenced by both their size and concentration. As the size of the CNTs decreases, their surface-to-volume ratio increases causing a stronger bond between the CNTs and the microorganisms and disruption of their morphology, metabolic processes, and cellular membrane [135]. Benigno et al. studied the antimicrobial activities of low-density polyethylene (LDPE)-based nanocomposites containing MWCNTs and confirmed that they were effective against DH5α *E. coli*. Their findings showed that the addition of 1% MWCNT filler to LDPE matrix provided synergistic features and produced a nanocomposite with outstanding mechanical and antibacterial capabilities. Additionally, these authors confirmed that the formation of biofilms increases *E. coli* adhesion to the surface of the materials, thereby reducing the antibacterial activities of MWCNTs against DH5α E. coli [136].

Light-activated antimicrobial agents deployed in surfaces

There are coatings developed that produce reactive radical species when activated by visible (white) light. Basically, these coatings comprise a photosensitizer (e.g., toluidine blue; methylene blue; rose bengal) immobilized in a material such as cellulose acetate. The activation of the photosensitizer results in the generation of radicals such as superoxide, the hydroxyl radical, and singlet oxygen. These reactive species directly attack the plasma membrane and other cellular targets in microorganisms, resulting in bacteriolysis [120,131]. This phenomenon forms the basis of "photodynamic therapy" (PDT) which is commonly used to treat a large number of skin cancers. More recently, PDT has been successfully used to reduce wound-associated MRSA. Coatings containing photosensitizers have been shown in laboratory studies to be effective against a wide range of microorganisms. Their effectiveness in the clinical setting has also been evaluated [137]. For example, in one of the studies, cellulose acetate coatings were placed under a fluorescent light and left exposed to a clinic ward atmosphere. After 24 h, it was found that significantly fewer bacteria were recovered from coatings containing a photosensitizer than from those that were photosensitizer-free. Also, a more recent study involved the coating of 15 computer keyboards. The keyboards were placed at randomly selected bed spaces within an ICU and sampled daily over a period of one month. In comparison to control (the non-coated) keyboards, the light-activated antimicrobial coating had a reductive effect on aerobic colony count. Although this effect was significant it was relatively small [120]. In hospitals, pathogenic bacteria frequently contaminate the privacy curtains. In light of this, it is possible to reduce curtain contamination by employing light-activated antimicrobial materials. However, according to regulations, a hospital's interior lighting requirements must range from 200 lux (in ward corridors) to 1000 lux (in accident and emergency departments) to 50 000 lux (in operating rooms). Therefore, it is important to moderate the usage of light-activated antimicrobials in all ward environments.

Mechano-bactericidal surfaces

The use of nanospikes for mechano-bactericidal applications is currently being explored [138,139]. This approach was inspired by the knowledge that nanospikes presence on the wings of dragonflies and cicadas can physically rupture bacteria on contact resulting in cell death [140,141]. This discovery offers a new way of fabricating nanostructured antibacterial surfaces, particularly for biomedical implants without the use of antibiotics or additive chemicals. Adopting this approach will significantly reduce biofilm formation since the cells are killed on contact and the bacteria cannot develop a protective response because the method is independent of the surface chemistry [142]. Aaron and co-workers showed the bactericidal activities of multi-directional electrodeposited Au nanospikes against *P. aeruginosa* and *S. aureus* bacteria as seen in Fig. 7(a) [143]. The biocidal actions of the Au nanospikes were estimated to be ~88%. Recently, Tian et al. [144] demonstrated 59% efficiency of silicon nanospikes against *E.coli* within 30 h of contact as shown in Fig. 7(c). Although the mechano-bactericidal effect has been attributed to the adhesion forces between the nanospikes and the bacteria cell [141], several other works have suggested the likelihood of other actions that depend on the stored mechanical energy, bacterial motility, and reactive oxygen species as shown in Fig. 7(b) [145,146]. In a study done by Valiei and co-workers, they showed that interfacial capillary forces can impact mechano-bactericidal actions [147]. Their initial experiment confirmed less significant mechano-bactericidal activities when *P. aeruginosa* was subjected to superhydrophilic nanopillars in an immersed moist environment. Surprisingly, the bacteria were shown to be physically ruptured and dead after the air/water interface was displaced occurring from water evaporation or trapped bubbles on the nanopillars, hence, confirming the effect of external capillary forces.. On the effect of capillary forces on nanospiked substrates with different contact angles, Valiei et al. recently proved that the superhydrophilic surface offers the highest mechano-bactericidal effect. The substrates showed a loss of mechano-bactericidal actions as hydrophobicity increases: the Laplace pressure on the hydrophobic substrates counteracts the surface tension forces thereby decreasing the total capillary force attraction [146].

Bacterial-repellent surfaces

Bacteria-repellent surfaces use the physical repulsion of bacteria cells from the surface of a substrate to stop the bacteria from adhering to the surface, thereby preventing subsequent biofilm formation. These repulsive actions occur without displaying any form of bactericidal effect. These repellent surfaces are normally designed using graft polymers covalently attached to substrates in close-knit ordered arrays. The polymer chains can be linear, branched, or cyclic, and are usually fabricated using oxygen-rich monomers that permit the chains to easily partake in hydrogen bonding [148]. The most commonly use polymer is Polyethylene glycol (PEG) because it is soluble in water, very flexible, nontoxic, biocompatible, possesses low immunogenicity, is endorsed for internal consumption, and achieves extremely large exclusion volumes. Its effective bacterial repulsion ability can be associated with the expanded hydration layer (therefore, a large excluded volume effect), a quick change in configuration, and steric repulsion [149]. Ionov et al. reported on surface-adsorbed Poly(aminoethyl methacrylate)/PEG copolymers [150]. Hui et al. worked on PEGylated PANI (PANI/PEG) nanofibers [151]. Wazawa et al. fabricated PEG monomethyl ether grafted to glass surfaces treated with (3-aminopropyl)dimethylethoxysilane and poly(acrylic acid) [152].

Besides PEG, some works have reported the use of polyamides, polyacrylates, and polysaccharides [149,153]. These polymers are hydrophilic, hydrogen bond acceptors, and electrically neutral. Therefore, the polymer chains are readily enclosed in a hydration shell of water molecules resulting in the formation of a strong hydrogen bonding network. As a result, each polymer chain is bound by steric interaction with all of its bordering chains. Every chain is somewhat slightly stretched out into an array of closely-knit springs normal to the surface of the substrate, and the whole polymer layer becomes swollen with water [148,153]. Along with bacterial repulsion, these polymer surfaces can also prevent the adsorption of proteins. In order to incorporate the surface hydrophilization agent, Ngo et al. used a number of techniques, such as physisorption, hydrogel network creation, surface grafting, layer-by-layer (LbL) assembly, and combining base polymers with surface modifying additives (SMAs) [154]. In 1993, Amiji and Park

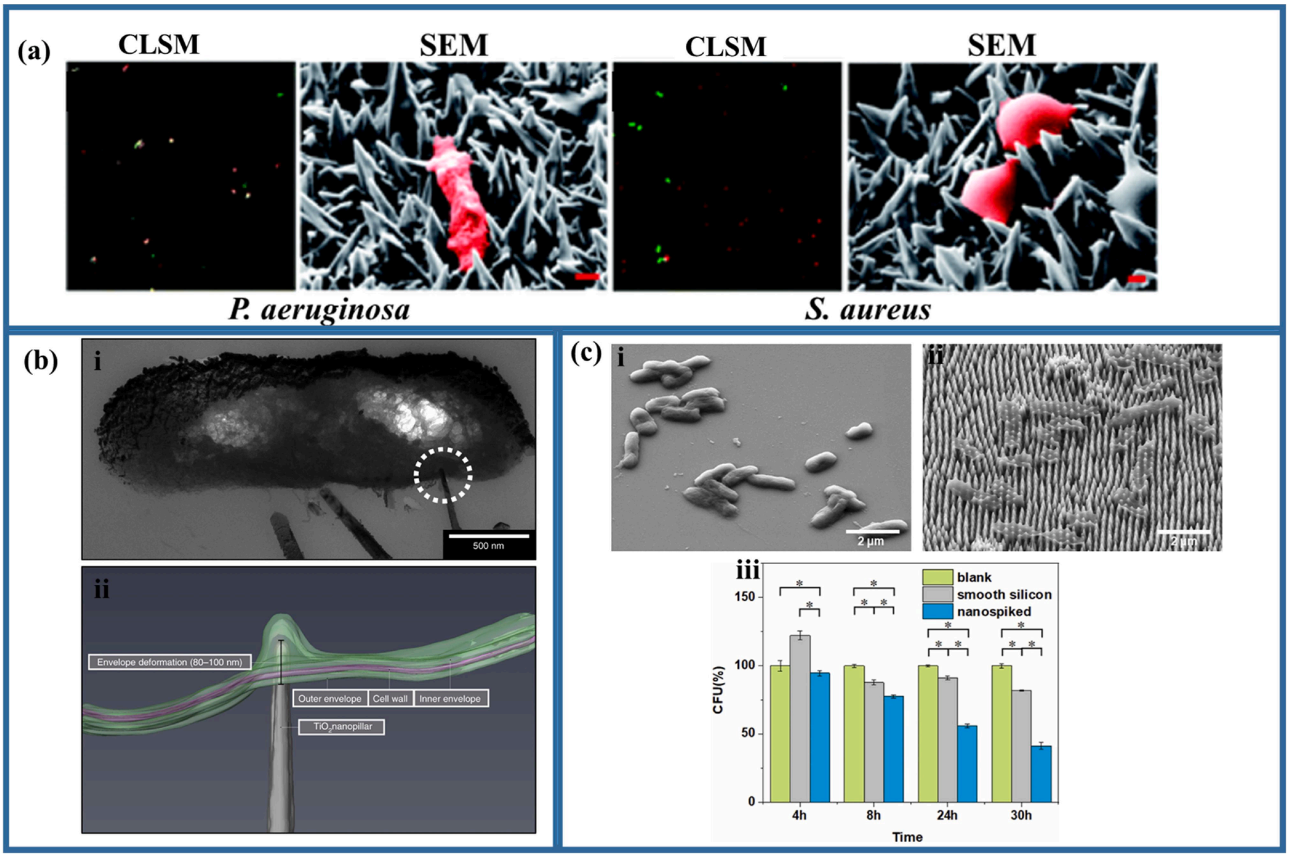

Fig. 7. (a) CLSM and SEM images of the bacteria atop nanospiked surfaces. The SEM images showed the trapping and rupturing of the bacteria. Live and dead bacteria are stained green or red, respectively in the CLSM images [143]. (b) TEM analysis showing a single nanopillar penetrating the envelope (dashed white circle) on *E. coli* (i) and Electron tomography showing the interaction of the *E. coli* cells with TiO_2 nanopillars suggesting the effect external forces (ii) [145]. (c) SEM pictures of *E. coli* on the; (i) smooth silicon and (ii) nanospiked silicon. *E. coli* bacteria on the smooth silicon showed intact cell shapes and cell multiplication whereas the *E. coli* bacteria on the silicon nanospikes exhibited ruptured cell membranes and cell death. (ii) Normalized plot of colony-forming unit (CFU) values at varying times. (B) [144].

reported that the grafting of biomaterials with either polyethylene oxide (PEO), albumin, or heparin is able to block platelet adhesion and plasma protein adsorption mostly through a steric repulsion mechanism [155]. In a work by Aghajani and Esmaeili, many protein/cell interactions that often control the effectiveness of biomedical devices were examined at the blood-biomaterial interface [156]. Deflorio et al. offered an interesting explanation regarding the antifouling mechanism; The bacteria cell must dispel the spring-like polymer chains and the surrounding water in order to attach to the surface of the substrate. This displacement can cause a rise in the osmotic pressure of the entrapped water-swollen polymer, presenting a work penalty that must be defeated before the bacterial cell can reach and attach to the surface of the substrate [148]. Notwithstanding the interesting properties of PEG, the techniques used to attach them to substrates are mostly complex and not economically viable. Besides, PEG is easily oxidized when biological materials are present, they are subjected to enzymatic cleavage reactions and fouling by cationic materials like lysozyme. Other limitations include inadequate mechanical durability and easy loss of efficacy [148,149].

Furthermore, zwitterionic polymers are also used to improve water interactions and the antibacterial characteristics of substrates. Zwitterionic polymers having covalently bonded anion and cation pendant groups and can electrostatically interact with polar water molecules leading to the formation of even more strongly bonded hydrogen networks [148]. A lot of zwitterionic substances are found in nature, including taurine in animal tissues, phosphorylcholine in cell membranes, osmolytes in saltwater fishes, and glycine betaine in plants [149, 157]. These substances inspired the intelligent fabrication of new ultralow fouling zwitterionic brush surfaces, such as polysulfobetaine, polyphosphorylcholine, and polycarboxybetaine (PCB) [149]. Zwitterionic antifouling surfaces are known to tightly bind water molecules, making them more stable than neutral hydrophilic polymer brushes through their electrostatically effected hydration [153]. Same as PEG, zwitterionic polymers adhere poorly to substrates and environmental changes can make them lose potency. Also, charge screening by ions can render them inefficient [148].

Multifunctional surfaces

Researchers have attempted to fabricate multifunctional surfaces, particularly surfaces that can kill bacteria (bactericidal surfaces) in combination with capabilities to prevent bacteria attachment and propagation (bacterial-repellent surfaces). These surfaces have the potential for improved antibacterial functions as shown in Fig. 8.

Xu et al. [159] reported strong bactericidal, biofilm-resistant, and virulence-suppressing potential of vitamin C (VC) on carbapenem-resistant hypervirulent *Klebsiella pneumoniae* (CR-hvKP). The results demonstrated that the bactericidal effectiveness is connected to the activation of reactive oxygen species (ROS) formation and dose-dependent in both the in vitro and mouse infection models. The VC also functioned as an inhibitor for biofilm formation of CR-hvKP by hindering exopolysaccharide (EPS) production. Additionally, at sub-minimum inhibitory concentrations (sub-MIC), VC performed as an efflux pump blockage and prevented the transport of EPS and capsular polysaccharide to bacterial cell surfaces, hence reducing the formation

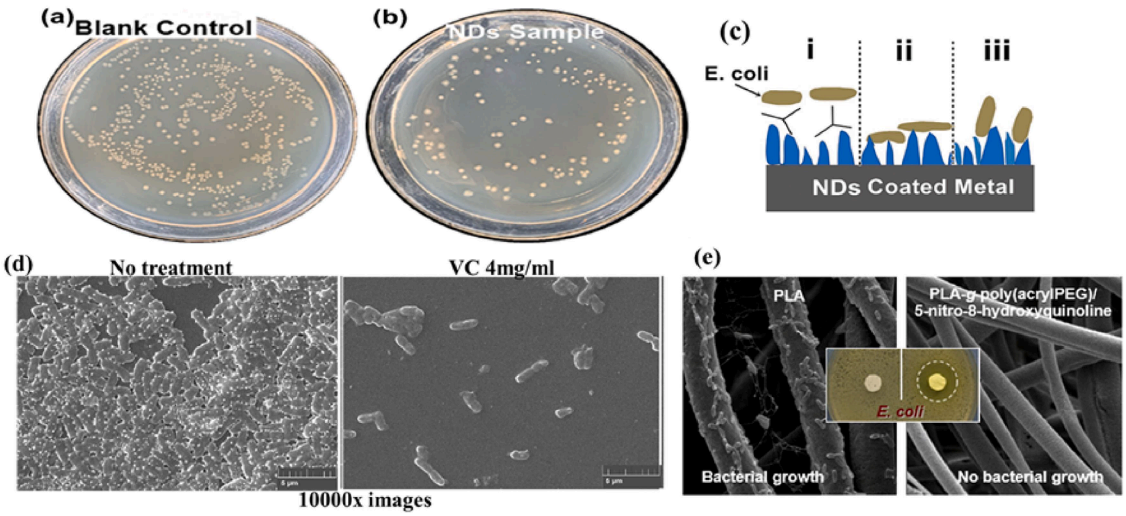

Fig. 8. Images of Petri dishes cultured with *E. coli* bacteria colony for 24 h for the (a) sample without the superhydrophobic surface and (b) sample with the superhydrophobic surface, (c) Schematic representation of the multifunctional surface; (i) bacteria resistant (ii) bacteria rupturing and (iii) bacteria trapping [158]. (d) SEM images of hypervirulent Klebsiella pneumoniae (hvKP3) biofilm formed on non-treated and VC-treated surfaces [159]. (e) SEM images showing the anti-bacterial capability of multifunctional polyethylene glycol (PEG)-based antibacterial [160].

of biofilm and capsule. In addition, virulence-associated genes in CR-hvKP were downregulated after exposure to sub-MIC VC. The findings proposed VC as an efficient and safe therapeutic agent for the treatment of CR-hvKP infections if the case is urgent or all current treatment options explored fail.

Uzoma et al. [158] showed that a superhydrophobic surface prepared with functionalized nanodiamonds and siloxane-acrylic resin can serve as effective bactericidal and biofilm-resistant agents. They confirmed that the presence of the low surface energy material and the narrow gaps/crevices on the coated surface will prevent bacterial attachment while the sharp edges of the nanoscale roughness can kill the bacteria via mechanical rupture. Similarly, superhydrophobic titanium-based medical implants have been reported to demonstrate strong multifunctional antibacterial actions [161].

In a work done by Li et al. [162] copper nanoparticles (CuNPs) were obtained by in situ reductions of Cu ions into a mussel-influenced hyperbranched polyglycerol (MI-hPG) coating by a simple dip-coating process, with coating possessing antibacterial and osteogenic characteristics. Because it has superior biocompatible properties and is easy to post-modify, an electrospun fiber membrane of polycaprolactone (PCL) was chosen as the substrate in this study. PCL electrospun fiber membrane is extensively utilized in embedded biomaterials for cardiovascular stents and bone regeneration purposes. In vitro antibacterial experiments against *S. aureus, E. coli*, and multi-resistant *E. coli* demonstrated the effective antibacterial performance of the CuNPs-added PCL membrane [PCL-(MI-hPG)-CuNPs]. Moreover, the in vitro results showed that PCL-(MI-hPG)-CuNPs' osteogenic properties were achieved via upregulating osteoblast-associated gene expressions and protein activity. CuNPs can be used to balance antibacterial and osteogenic qualities in a surface coating, according to this study. Using mussel-inspired catechol chemistry, Ye et al. [163] obtained a new macromolecular framework with thin-film composite (TFC) membranes and block copolymer brushes with dual functionalities for combined "defending" and "attacking" tactics against biofouling.

Activators regenerated by electron transfer–atom transfer radical polymerization (ARGET-ATRP), an eco-friendly and controlled polymerization technique, were used to graft quaternary ammonium salt (QAS) polymeric brushes possessing bactericidal potential and zwitterionic polymer brushes possessing high capacity for hydration on TFC membranes. Considering the TFC membrane modified, the film displayed reduced biofouling tendency and good bacterial inactivation.

The literature has reported several materials as being useful when combined with other materials to obtain bactericidal and bacterial-repellent multifunctional surfaces [164–166]. Polyethylene glycol (PEG) has demonstrated good potential as an anti-adhesive (bacterial-repellent) component in surfaces with bactericidal and bacterial-repellent capabilities [167–169]. For instance, Wang et al. [169] created a coating material with dual functionality that is both anti-biofouling and self-healing. With a triple-crosslinked network (covalent connections, hydrogen bonds, and ionic interactions), this substance could be coated on 316 L stainless steel substrates and displayed strong adhesive strength. PEG served as the component that was bacterially resistant. Additionally, the surface's characteristics might be affected by the PEG inclusion process. Moreover, polylactide (PLA) and polyethylene glycol (PEG)-based antibacterial micro- and nanofibrous materials were created via electrospinning [160]. In connection to mats application, investigations were conducted into how the inclusion process affected surface wettability, thermal, mechanical, and biological characteristics. They did have a plasticizing effect on PLA, which was more pronounced in the case of the physical blend fibers and related to the higher PEG mobility. Neither physical blending nor chemical grafting had an impact on the mats' wettability. The PEG inclusion method also had an impact on the mats' mechanical qualities. Mats made by physical blending of PLA and PEG were discovered to be quasi-ductile, but mats made from chemical grafting of poly (l-lactide)-graft-polyethylene glycol methyl ether acrylate were discovered to be brittle. As a model antibacterial medication, 5-nitro-8-hydroxyquinoline (5N8Q) was encapsulated. For all PEG-containing mats, the polyether inclusion method influenced the release patterns of 5N8Q, with a burst effect noted. Furthermore, all drug-loaded mats demonstrated antibacterial efficacy against the activity of *E. coli* and *S. aureus* as observed in Fig. 9(e). As a result, the method of polyether inclusion chosen is determined by the antibacterial materials' intended application [160]. Other materials have also proven effective as bacterial-resistant components on surfaces with both bactericidal and bacterial-repellent capabilities. They include polysulfobetaine methacrylate, PSBMA; [163] poly (2-methacryloyloxyethyl phosphorylcholine), PMPC; [170] poly(N-hydroxyethyl acrylamide), PHEAA; [171] functionalized silica nanoparticles, F-SiO_2, [172] polydimethylsiloxane, PDMS; [173] Poly (acrylic acid), PAA; [174] lysine methacrylamide, LysAAm; [175] heparin, HEP; [176], etc.

In a similar manner, a number of materials have been explored as bactericidal components in multifunctional surfaces [180–182]. For example, through electrostatic interactions, PEB grafted on a

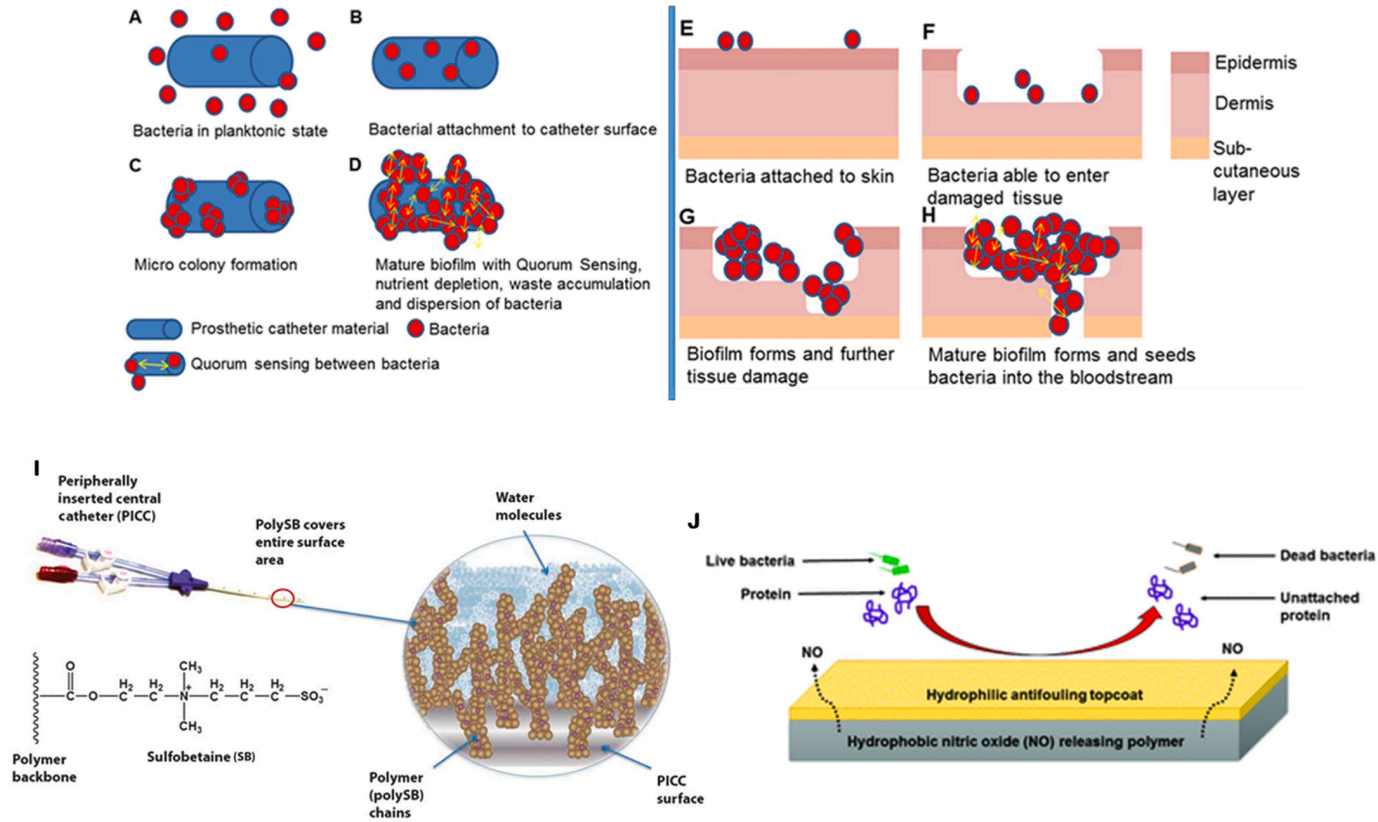

Fig. 9. Biofilm contamination of catheters and wounds. A–D depict the colonization stages of a catheter with the attachment of plank-tonic cells, generation of micro colonies, and the maturity of biofilm. E–H displays a normal human skin barrier colonized with bacteria from the flora prior to an injury permitting bacteria to reach lower layers, thereby causing damages to the deep-lying tissues, and ultimately infiltrating the bloodstream. Adapted from Hughes and Webber [177]. (I) Scheme for fabricating an antibacterial surface on a medical catheter surface [178]. (J) Action mechanism of antifouling polymer containing Nitric oxide (NO) [179].

polydimethylsiloxane (PDMS) network is found to kill bacteria [183]. Triclosan has demonstrated effectiveness as a bactericidal component when used in a supervised release approach in slippery liquid-infused porous surfaces (SLIPS) to prevent microorganisms from fouling surfaces and to get rid of non-adherent pathogens in surrounding media [184]. By cross-linking chitosan-g-eugenol/zwitterionic copolymer on the electrospun membranes to fabricate unified antibacterial and antifouling surfaces, Li et al. showed chitosan as a bactericidal component [185]. Other bactericidal components in the literature include silver nanoparticles (AgNPs), [186] poly[2-(methacryloyloxyethyl] trimethylammonium chloride (PMETA), [163] polyhexamethylene biguanide (PHMB), [187] graphene oxide (GO), [188] quaternary ammonium salt polymer (QAC), [169] chitosan (CS), [176,189] lysozyme, [190] polycarbonate, [176] etc.

Most recent techniques and applications of antibacterial surfaces

Biomedical application

While there are over 2 million infections associated with healthcare each year taking the United States as an example, only about 100,000 of these cases lead to death [191,192]. Similar statistics from China's National Health Commission indicate that infections brought on by microorganisms are responsible for 29.3% of cases of food-borne diseases [193]. Device-related infections (DRIs) associated with implant surgical procedures and temporary biomedical devices have become popular with the increasing applications of biomedical devices in healthcare systems. Unlike temporary devices that are regularly replaced to minimize infection threats, implant surgical procedures present more challenging solution routes as the implant surfaces have been proven to be mostly prone to biofilm formation due to the adhesion and multiplication of several bacteria. Infection is considered as the outcome of the complex relationships between biomaterials and human hosts. The immune systems of the hosts are usually expected to efficiently purge all opportunistic pathogens capable of introducing contaminations. However, for implant-based DRIs, acute and chronic inflammations result from the triggering of local tissue responses. This resultantly causes reactions on the foreign body, thereby encouraging microbial colonization on the implant surface [192]. Due to bacterial drug resistance, these illnesses are extremely challenging to treat [194]. The three main phases of microbial contaminations are: the contaminations that occur during surgical procedures which often results to prosthetic contamination after surgery; the subacute infections which emerge within 3–24 months sequel to surgery; and the asymptomatic late infections phase which typically occur after 24 months [194]. Generally, microorganisms that are gram-positive and gram-negative are mostly implicated in implant-based DRIs.

One efficient approach to prevent these threats in biomedical applications is the development of antibacterial surfaces. Bacteria are the most common biofilm-forming microorganisms, and at least 95% of most bacterial populations are capable of film formation [195]. They easily colonize surfaces of implants, vascular stents, and other biomedical devices, and can insulate themselves with protective biofilms. The growth and development of bacteria occurring on the surface of catheters and wounds on normal human skin were adequately described by Hughes and Webber [177] as illustrated in Fig. 9. Antibacterial surface coatings are employed to deplete or minimize bacterial colonization, thereby reducing biofilm development on the said surfaces. They function by either adjusting the topography of the surfaces or by chemically modifying the surfaces; hence they exhibit antifouling and bactericidal effects [196,197].

The Antifouling effects constitute both fouling-resistant and fouling-release approaches, which basically involve the prevention of bacteria attachment and reduction of bacteria adhesion, respectively [198–200]. As such, many superhydrophobic [201,202], superoleophobic [203, 204], and liquid-infused porous surfaces [199,205,206], as well as hydrogels [207], have been reported for their antifouling activities. The bactericidal effects incorporate the utilization of germicides and antibiotics [208,209], ion-releasing metals [210,211], peptides [212,213], cationic polymer chains that are capable of eliminating attached bacteria [214,215], quaternary ammonium compounds [216,217], and so on. Based on the applied mechanism for bacteria killing, bactericidal surfaces can be viewed in two categories: (I) contact-based, and (II) release-based bactericidal surfaces. The contact-based bactericidal surfaces are fabricated by physisorption or covalent conjugation of antibacterial agents (like polycations, quaternary ammonium compounds, etc.) which directly kill attached bacteria, while the release-based bactericidal surfaces involve preloaded biocides that slowly released into the surrounding environment to activate bacteria killing. Moreover, some bactericidal surfaces contain two distinct biocides integrated into a single system and they adopt both mechanisms for contact-based and release-based surfaces in their operations. These are generally called dual-contact- and release-based bactericidal surfaces. Smith and co-workers fabricated an anti-bacterial coating on the lumen and surface of catheters via van der Waals force, by employing polyelectrolyte /polyelectrolyte-surfactant complexes [178]. This coating can also be made on equipment of different sizes and shapes, and they possess broad-spectrum anti-bacterial properties and good biocompatibility. Catheters fabricated using this technique possess good anti-bacterial colonization potential and reveal durable antibacterial characteristics in vivo. Fig. 9(I) gives a vivid pictorial account of the construction of antibacterial surfaces on medical catheter surfaces.

The last few decades have witnessed the emergence of nitric oxide (NO)-based therapies as a potential bactericide and antithrombotic substitute to the present state-of-the-art methods to stop pathogenic infections and clot formation in implantable devices [218–220]. NO is an endogenous gas molecule that performs various major physiological actions which include preventing platelet adhesion and activation, suppressing adhesion and proliferation of bacteria, improving vasodilation, promoting angiogenesis, and assisting wound healing [221,222]. The antibacterial efficiency of NO has been demonstrated against infection-causing agent pathogens such as *P. aeruginosa, E. coli, S. epidermis, S. aureus, Acinetobacter baumannii,* L. *monocytogenes, and E. faecalis* [223]. The antimicrobial mechanisms of NO include nitrosation of amines and thiols in the extracellular matrix, lipid peroxidation and tyrosine nitration in the cell wall, and DNA cleavage in the cellular matrix [224]. To simulate the physiological release of NO from the endothelium, several NO donors such as diazeniumdiolates and S-nitrosothiols have been constructed and can be integrated into polymeric materials (Fig. 9(J)) for localized delivery of NO [225]. The Meyerhoff group at University of Michigan, and the Handa and Brisbois groups at University of Georgia are at the forefront of this research; they have reported significant progress [179,223,226-228].

Another strategy for fabricating bactericidal surfaces in medical applications is the fabrication of bacteria-repellent surfaces. These are antibacterial surfaces that limit the initial extent of bacterial adhesion, hence obstructing biofilm formations at the earliest stages. The surfaces often generate physical barriers called hydration layers by undergoing improvements with hydrophilic polymers, and they are grouped into two categories according to the mechanisms of formation of the layer, namely zwitterion-based [229,230] and ethylene glycol-based surfaces [217,231]. As opposed to the actions of bacteria-repellent surfaces, antibacterial surfaces for medical applications may be designed with bacteria-release properties. While these surfaces permit the initial clinging of bacteria, the adhered bacteria are released under certain conditions or environmental stimuli [232]. These may be thermo-responsive [233], pH-responsive [234,235], or some other unconventional external impulse, such as changes in mechanical stretching and electrical voltage [236,237].

Food processing and packaging applications

Food contact surfaces (FCS) consist of all surfaces that may come into contact with food products during production, processing, and packaging operations. These surfaces are made from a range of food-grade and/or food-safe materials like stainless steel, ceramics, plastic, glass, rubber, or wood. The materials must fulfill the requirements specified by agencies such as US Food and Drug Administration (U.S. FDA, 2007) or European Food Safety Authority (EFSA, 2008), or other national equivalents. They should be nontoxic, nonabsorbent, easy to clean and maintain, and are not expected to leak any substance into the food product. Also, they should possess smooth surfaces without microholes, cuts, and crevices that might enable the build-up or trapping of biological material. For instance, scientists have recently shown that exposure to some Per- and poly-fluoroalkyl substances (PFAS) in the environment may be toxic to humans and animals. PFAS have been used to provide oil and water resistance to paper products for both food and non-food applications [238]. Perfluorooctyl sulfonamide ethanol-based phosphates were the first substances employed as grease repellence in food contact papers, followed by fluorotelomer thiol-based phosphates and polymers [238]. Even though PFAS have been in use for more than eight decades, scientific understanding and technical equipment required to test for PFAS at very low concentrations in food just started not long ago. In 2016, FDA disclosed that it has stopped providing for the use of certain PFAS in food contact paper and paperboard [239]. In another case, they also disclosed that it has stopped providing for the use of certain perfluoroalkyl phosphates and acrylate copolymers in food contact paper and paperboard [240]. DeFlorio et al. provided a broad list of everyday approved FCS and their specific applications [148].

The possibility of food contact surface (FCS) contaminating with various microorganisms for example *S. aureus, Salmonella enterica, E.coli, and* L. *monocytogenes* in food processing plants is very likely [12]. Over time, different studies have been carried out on the use of antimicrobial treatment of FCS, and many classifications have been outlined based on the surface properties, bacteria species, treatment required, and legal framework [241–245]. Also, a large amount of research has been devoted to the use of active packaging (i.e. use of antibacterial packaging materials) to prevent the susceptibility and vulnerability of food to any form of microbes and to effectively extend the food shelf-life [246, 247]. The principle of active food packaging is based on treating/adding to the matrix of the FCSs an active antimicrobial component, which adhered to them [248] or coating the surfaces with the appropriate functional coatings by modifying surfaces to endow packaging materials many functional activities, for example, bacterial control and oxidation resistance [249]. Compared with crude or traditional packaging, active packaging eliminates the need to directly introduce active ingredients into food where not only the original flavor is maintained but additionally mitigates bacteria and prevents spoilage, in order to ensure elongation of shelf-life [246,250]. Some of the generally used active antimicrobial agents include Ag nanoparticles, ZnO, CuO, Chitosan, essential oils, etc. as shown in Table 2. Essential oils are individual volatile compounds extracted from aromatic plants and are Generally Recognized as Safe (GRAS) [251]. The essential oil can inactivate essential enzymes, induce coagulation of cytoplasm, disruption of genetic material, and eventually affect cell viability [252]. Current research involves the encapsulation of essential oils in micro/-nanocontainers to achieve effective antibacterial actions [253]. Examples of essential oil include Oregan, Clove, Rosemary, Cinnamon, Eucalyptus radiate, methyl chavicol, etc. Major drawbacks normally encountered in the use of antimicrobial agents include; high cost of production, high intensity of off-flavors (foul odors), and inability to withstand intensive processing conditions (Many potential agents

Table 2
Examples of antimicrobial agents used in Food packaging together with their matrix material and target microorganisms.

Agents	Matrix material	Target Microorganisms	References
Inorganic Antimicrobial Agents			
Ag ions and Ag nanoparticles	Polyethylene, Polyurethane, Poly (ethylene terephthalate), cellulose acetate, Hydroxypropyl methylcellulose, PDMS, polyvinyl alcohol, zein corn protein, tragacanth, pectin, gelatin, and pullulan	*Bacillus subtilis, E. coli, S. aurus,* L. *monocytogenes, Salmonella enterica*	[254–256]
ZnO	Starch, Alginate, Polylactic acid, Glass, Chitosan	*Candida albicans, S. Typhimurium,* Gram-type bacteria	[257–259]
CuO	Polyethylene, Nylon, Polyactic acid	*S. aurus, E. coli,* L. *monocytogenes, P. aeruginosa, Salmonella enterica*	[260–262]
Organic Antimicrobial Agents			
Synthetic antibacterial agents			
Vanillin	Chitosa, Polyvinyl alcohol	*Cronobacter Sakazakii, Salmonella enterica, E. coli, S. aureus, P. aeruginosa*, fungal inhibition	[263,264]
Phenols	Chitosa, Polyvinyl alcohol, Starch, Alginate, polypropylene, low density polyethylene	*E. coli, S. aureus, P. aeruginosa*	[251,265,266]
Quaternary ammonium salts	Chitosan, Polyvinyl alcohol	*Klebsiella pneumonia, Enterococcus faecium, Acinetobacter baumannii, S. aureus, P. aeruginosa,* and Enterobacter species	[267,268]
Natural antimicrobial agents			
Chitosan	Polyethylene and polypropylene	*S. aureus, Salmonella, Enterococcus faecalis,* L. *monocytogenes*, and Gram-type bacteria	[269,270]
Lysozymes	Pectin, Chitosan	*S. faecalis and E. coli*, Gram-type bacteria	[271,272]
Essential oils	Polyethylene, Polyvinyl alcohol, β-cyclodextrin, Alginate, Galbanum gum, Chitosan, Starch	*E. coli, Enterococcus faecalis, P. aeruginosa, K. pneumoniae, Acinetobacter baumannii, and Salmonella Typhimurium, Bacillus cereus*, etc.	[148,273,274]
Nisin	Hydroxypropyl methylcellulose, Chitosan	L. monocytogenes, E. coli, B. cereus, Clostridium perfringens	[275–277]

denature at high temperatures and pressure). The two modes of active packaging will be analyzed in detail in this section.

Use of antibacterial or antioxidant agents

This method uses the phenomenon of adding substances like antibacterial and/or antioxidant agents inside or on the surfaces of materials used food packaging. The general intended purposes are classified into three, which are; improving the surface characteristics of food packaging, cleaning the surfaces, and adding antimicrobial capabilities to the surface [12]. Because it is inexpensive and meets the hygienic requirements for food packaging, polyethylene film (PE) is widely employed in daily life as an antibacterial agent [278]. Despite the fact that PE films make an effective moisture barrier that prevents external bacterial pollution, they lack an active capability and are unable to address the issues of bacterial proliferation and food oxidation in food [279]. Based on this deficiency, researchers continued in further investigation to develop or detect a better antibacterial agent, of which low density polyethylene (LDPE) was discovered. LDPE became common industrial polymer overtime and is commonly used in food packaging for because of its useful properties which include its ease in shaping, handling and reusing, and its high-cost efficiency [280–282]. Also again, after several usages, it was found that LDPE lacks proficiency in some of its intrinsic traits such as printability, adhesion to the surface, and some other defects in surface characteristics such as inert surface with extremely low surface free energy (wettability) [283]. Table 3 presents a categorization of antimicrobial agents applied in FCS based on their mode of application (intentionally and unintentionally added) and their specific uses.

Use of antimicrobial coatings (AMCs)

In the quest of developing good practices in preventing food products and food contact surfaces from microbial contamination, AMCs were developed and adopted by several food standard agencies of different countries as an outstanding approach. AMCs must conduct functional and utilize metrics as well as several strict regulatory requirements in order to be used in real applications. When used in a complicated environment, the perfect AMC is potent, stable and resistant to chemicals, harmless, affordable, and simple to clean [285]. Nevertheless, most AMC work has been on enhancing the coatings' antibacterial characteristics in order to realize this concept. There are different techniques for coating AMCs, which are:

Electro-deposition: In this process, the requirement is for the coating material to dissolve in ionic form allowing for their migration to the surface, where they create the coating as a result of a reduction-oxidation reaction started by an applied voltage [286]. The coating is extremely adjustable because it is made of dissolved ions and is managed by an applied voltage.

Vapor deposition and atomic layer deposition: This process depends on gaseous precursor molecules that are highly reactive being displaced [287]. When the gaseous precursor reaches the substrate, it reacts quickly with the reactants that have already been deposited. The reaction results in the creation of a one-molecule thick coating on the substrate. Repeating the procedure results in consistent and fine coatings, albeit the coating procedure can take several hours when somewhat thick coatings are required.

Sol-gel method: Precursor chemical reactions are involved in this, which is similar to the vapor deposition process in the liquid state [288]. However, because the liquid components must first react until the required microstructure in the gelled mixture is accomplished, the sol-gel process does not allow control of the coating thickness at the molecular level. After that, the substrate is coated with the gel using dipping or spinning techniques. Thus, to create the final coating, the coated substrate is heated at a high temperature for a period of time (often between 500 °C and 1000 °C) [289].

Table 3
Classification of antimicrobial agents applied in FCS, their mode of application, and their specific uses.

FCS application	Antimicrobial presence in food	Antimicrobial regulatory classification	Example of antimicrobial	Example of surface
Decontamination of the surface	Unintentionally added	Food contamination or residue	Peracetic acid [283]	Countertops, table tops, cutlery, cookware, dishware, and other equipment for processing food decontaminated using antimicrobial agents
Food packaging	Intentionally added	Food additive	Silver ion [284]	Containers for bulk food storage, cartons, paperboard, jars, and bottles having an antibacterial effect

Five principles—anti-adhesion, antimicrobial-loaded, contact inactivation, photocatalytic, and multifunctional—cover the full spectrum of interactions with microorganisms that result in the prevention of bacterial proliferation on FCS and can be used to categorize the potential applications of AMCs [290]. These will be explored briefly.

Anti-adhesion: The fundamental idea behind anti-adhesion is the modification of FCS in a way that the microbes are driven away from the surface. The surface alteration may take the form of applying polymers like polyethylene glycol [291] or carboxybetaine polymer [292], or it may involve changing the surface topography [293].

Antimicrobial-loaded: An antimicrobial agent that has been included into a coating matrix must disperse into the food and/or environment in order to be effective. In the case of a simple release, the diffusive process might happen randomly, but it can also be triggered by an external stimulus. Antibacterial agent diffusion often depends on time in a simple release mechanism whereas it depends on stimuli in a stimulation sensitive mechanism. However, instances where just the bactericidal agent diffuses, and the coated matrix stays on the substrate have also been documented [294].

In contrast to the stimulation-sensitive AMC agent, which is only mobile when specific conditions are met, the AMC agent depletes more quickly in the simple release situation. Atefyeka et al. [294] developed a highly porous titanium oxide coating on SS in which gentamycin, vancomycin, and daptomycin were incorporated to stop post-surgery infections following the placement of medical equipment. The antimicrobial effectiveness was shown against *S. aureus* and *P. aeruginosa*. On the other hand, titanium was coated with polypeptides that were intended to release in reaction to the number of blood components present [295]. Both AMCs were intended for medical implant application.

Contact inactivation: The contact inactivation phenomenon is based on the harm that some long-chain compounds can do to microorganisms when the associated active molecules come into touch with the microorganism. According to this theory, the coating's active ingredient needs to be in contact with the surface to function as an antimicrobial. Murata et al. [103] referred to their coating as a polymeric brush that breaks down *E. coli* cell walls because of surface charges rather than the length of the polymeric chain employed to make the molecular brush. Glinel et al. [296] also referred to their coatings as polymeric brushes that, as a result of the inclusion of magainin I inside the brushes, exhibit antimicrobial activity and prevent biofouling. Tests on these coatings against *Listeria Ivanovii* and *Bacillus Cereus* were successful.

Photocatalytic: AMCs with a photocatalytic foundation seek to address the issue of active ingredient depletion. Because of the rapid rate at which it produces reactive oxygen species, TiO_2 is the key component of photocatalytic AMCs [297]. In order to produce hydrogen peroxide (H_2O_2), which is what gives TiO_2 and the other photoactive semiconductors like zinc oxide (ZnO) and magnesium oxide (MgO) their antibacterial properties, both a source of water and UV light must be present at the same time.

Multifunctional: The working principle is the combination of all the functionalities mentioned above to obtain a single AMC can enhance the effects of separate operating principles. For instance, Whitehead et al. [293] modified the surface topography of Ag coatings to reduce cell adhesion while killing L. *Monocytogenes, E. Coli, and S. aureus* by releasing silver ions. In another instance, the effectiveness of chitosan released from a photocatalytic ZnO coating against *Salmonella enterica, E. coli, and S. aureus* was tested [298]. Multifunctional coatings are more difficult to make, despite being a wonderful idea in theory.

Marine industry applications

The antibacterial surface will go a long way in dealing with the problem of contamination/fouling of micro- or macro-organisms in the aquatic/marine industry [299]. Marine biofouling is the unwanted buildup of marine microorganisms, flora, and fauna on surfaces of materials that are submerged, and it has a significant detrimental effect on the machinery and body parts in marine-based enterprises as seen in Fig. 10 [300–302]. A significant buildup of biofouling on marine vessels can be a daunting task. Traditionally, biocides, a chemical substance or microorganism capable of regulating the growth of harmful organisms by biological or chemical means, are used to mitigate marine biofouling. Marine biofouling makes ship hulls heavier and rougher, which results in frictional resistance and leads to increased fuel consumption. Additionally, it starts or speeds up the corrosion of metal and concrete structures, increasing the danger of failure of marine infrastructure and equipment [43,303,304].

Seawater pipes used in near-sea industries will become bio-fouling-prone, as will net cages employed in the aquaculture sector, which will reduce both the productivity of aquatic products and the equipment's efficiency [50,306,307]. Additionally, the organisms that cause biofouling on maritime vessels will move to different oceans that they do not naturally belong to and disrupt the ecosystem. Hence, marine biofouling represents a major concern that must be addressed for both the marine economy and the marine environment. The antibacterial surface technique can be applied to this problem to reduce marine biofouling build-up by using self-polishing paints, in which the biocide is released gradually when seawater comes in contact with the surface layer of the paint. Recently, copper-based antifouling paints have been used because they are less toxic in aquatic environments [308,309]. Also, the use of non-stick coatings without biocide addition but self-cleaning enabled surfaces which prevent fouling has been explored [158]. Motivated by the soft coral surfaces' effectiveness at preventing fouling, Bing et al. [310] fabricated an antifouling coating of graphene-silicone elastomers (GSE) composite materials having tentacle structure (TS-GSE). The designed film can dislodge bacteria via physical interaction and was found to be more efficient than bactericidal coatings. They assessed the bacteria attachment under both static and dynamic, and their findings proved that the TS-GSE film provided effective anti-adhesion properties for both Gram-positive and Gram-negative bacteria.

Furthermore, the most critical issue for the equipment and infrastructure used in the marine environment is always corrosion of materials [50,306]. Microbiologically influenced corrosion (MIC) connotes the deterioration of metallic materials that is hastened either directly or indirectly by the metabolites of microorganisms [311]. In the marine business, MIC is to blame for the majority of financial losses. Statistics show that around 20% of all economic losses are attributable to MIC [43, 312-314]. MIC is typically produced by anaerobic sulfate-reducing bacteria (SRB) and aerobic iron-oxidizing bacteria (IOB) [43]. In real field situations, by working together, these two microorganisms speed up the deterioration of materials. IOB reduces the oxygen content of the medium to create the perfect condition for anaerobic SRB growth, which in turn promotes SRB's matrix corrosion. Together, SRB and IOB carry out this process to build biofilms on metal surfaces, which are frequently composed of sessile cells, extracellular polymeric substances (EPS), and corrosion byproducts produced by these two bacteria [306]. A crucial part in MIC is played by biofilm [54]. For instance, in ship hulls, biofouling leads to the breakdown of coatings, increased drag resistance, and increased corrosion rate [315]. Antibacterial surfaces technique can be applied via the application of a multifunctional organic coating. Additional antibacterial effects are produced by the leaching of the hydrogel species incorporated in the coating matrix in response to biofilm activities [316].

Antibacterial surfaces and their weakness

Biocide-based antifouling (AF) coatings

Biofouling on surface coatings limits the performance of coated installations submerged in wet environments [317]. And biocides-based antifouling coatings have been and remained the primary strategy for

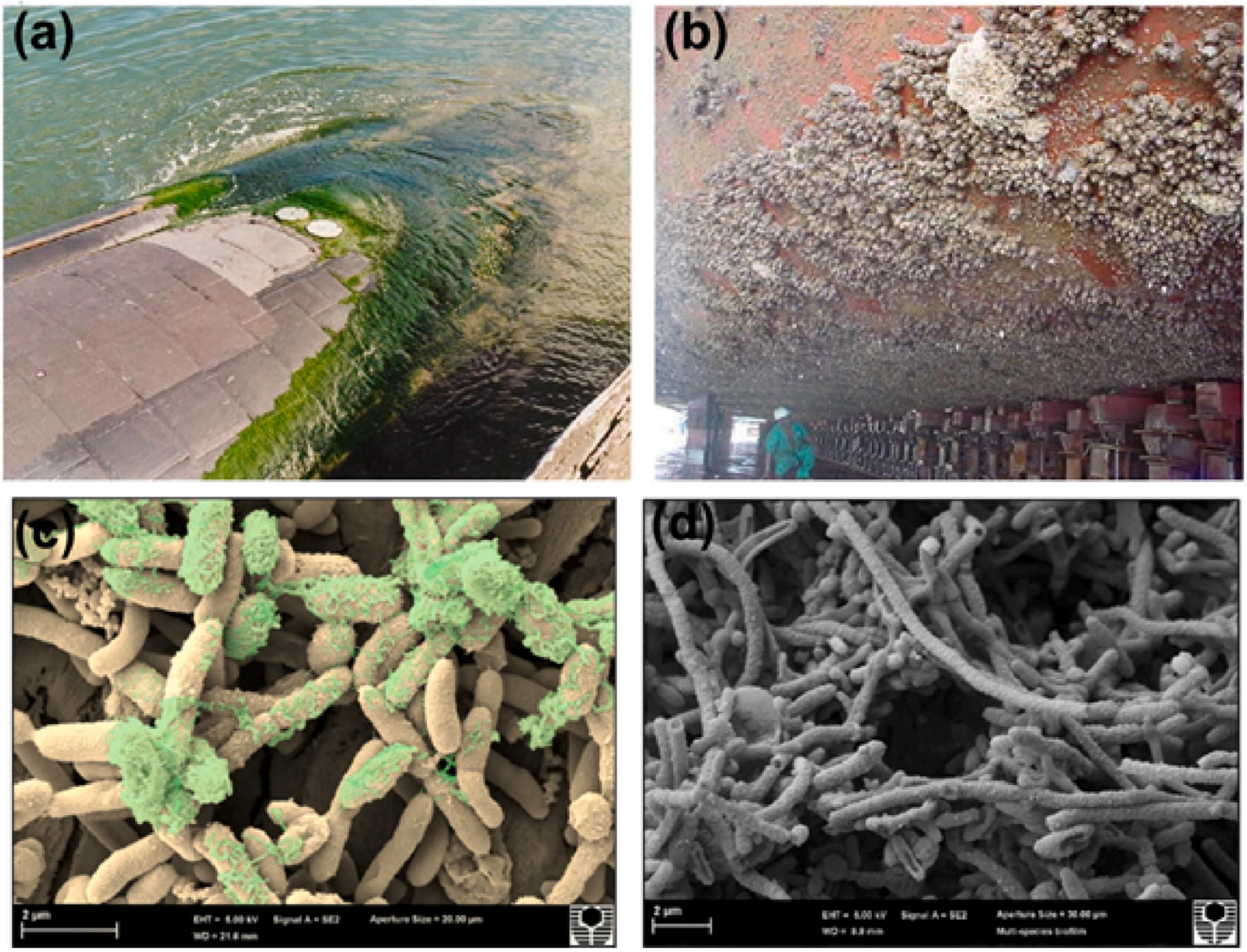

Fig. 10. Images of vessels fouled by marine organisms (a) green alga (seaweed) and (b) barnacles [17]. (c) SEM images of the formation of Klebsiella pneumoniae biofilm on carbon steel in artificial seawater and (d) FESEM image of biofilm formed by multiple species on carbon steel showing the complexity and close-knit living arrangement of multiple species marine biofilms [305].

compacting biofouling in marine and offshore industries [2,318]. Even now, biocide-based AF coatings are still the leading coating for controlling biofouling in marine and off-shore platforms. But then, the major drawbacks against the existing biocides-based AF coating systems are (a) the specificity in action (limited target organisms), (b) uncontrolled discharge of biocides into the environment which could interfere with the ecosystem by attacking non-target organisms, or which could become a localized source of organic biocide in the water bodies [318, 319], and (c) difficulties in incorporating the biocides in active form into many coating formulations due to possible interaction with the coating resin and/or the deactivation of the biocides in the coating resin. Consequently, therefore, in some scenarios, marine vessels and other related installations are still allowed by the growth of algae, barnacles, slime, etc., due to the lack of a multipurpose/broad spectrum AF coating system with minimal negative impact on the ecosystem [320]. Some reports, however, have reported achieving minimal toxic or eco-friendly biocide-based AF coating systems by: (a) selecting biodegradable biocides from natural sources [321], and (b) entrapping the biocides in nano/micro containers as ''trapped'' before embedding in the composite AF coating formulation [322]. Although these reduced the toxicity, some attacks on non-target organisms are still eminent, and long-term protection is not guaranteed except upon constant replacement which is not cost-effective.

Fouling release polymers

Fouling-release coatings (FRCs) are coatings that have surface-free energy which possess weak tolerance to the adhesion of microorganisms so that they can easily be removed by the water shear force. This technology has enjoyed popularity in the last decade because it is biocide-free and thus environmentally friendly. However, they are confronted with the obvious poor adhesion to substrates, mechanical strength, and low anti-fouling properties, which limit their applications. Hu et al. reported fouling-release coatings for potential marine antifouling applications that are based on silicone [323]. R.L. Townsin et al. reported on fouling control coatings that employ low surface energy and foul release technology [324]. The report revealed the physical and chemical properties of such coatings and the efficiency of surface smoothness in preventing fouling organisms from adhering to it [324]. Silicone-based coatings with superhydrophobic properties have been well reported in this regard. According to a new protocol described by Zhang et al., long-chain organosilanes react with water in a single step under regulated conditions to form hierarchical siloxane aggregates that may be dispersed in industrial solvents as the coating mixture [325]. The results reveal excellent superhydrophobicity exceeding 170° which can be achieved on varied materials (substrate) by simply spraying any coating material with the new organosiloxane mixture. Water-repelling

Table 4
Summary of the types of antibacterial surfaces with their advantages and limitations.

Anti-bacterial Surface	Description	Advantage	Limitation	Reference
Antifouling Paints	The paints often contain biocides	Effective in killing organisms and preventing fouling process	Short life time performance due to uncontrolled leaching and are non-selective against target organisms, hence, are not environmentally friendly	[328, 329]
Protein Resistant Polymeric Biomaterials	increase surface hydrophilicity and resist protein adsorption.	Effective in preventing the desorption of microorganisms	Can increase the water retention in coatings. They typically swell in marine conditions, which results in poor mechanical qualities and restricts their use.	[154]
Fouling-release coatings (FRCs)	Do not prevent organisms attachment, such as with antifouling and protein resistance, but ensures that the interfacial bond between the surface of the coating and the organisms is weak as a result of their low surface free energy (SFE), as this will make it easier for the ship's navigation or mechanical cleaning methods to remove the attached organisms.	Effective against foulers adhesion, and are eco-friendly with drag reduction and chemical stability	Weakens adhesion of the coating on substrates and general mechanical property	[323]
Switchable Antimicrobial and Antifouling Smart Coatings	pH variation acts as an external trigger for hierarchical rearrangement.	Effective against foulers adhesion, and are eco-friendly with drag reduction and chemical stability	Complex fabrication. Might weaken adhesion and mechanical properties	[170]

surfaces found application even in food packaging. Superhydrophobic coatings can prevent food packaging from becoming fouled or contaminated, reducing food waste and enhancing the shopping experience. Mahmut Ruzi et al. [326] reviewed the various materials that have found application in food package materials. Superhydrophobic coatings can enhance energy efficiency in the marine industry via the reduction of surface friction that occurs on ship hulls. The materials for achieving these are mostly inorganic materials such as TiO_2 and SiO_2-based materials. In all, the setback of this technology is the weak adhesion and mechanical properties.

Advances in smart antibacterial surfaces

Modifications have been made to achieve smartness in the mode of action of some antibacterial surfaces. Xu et al. [170] identified a reversible pH-induced transition between antimicrobial and antifouling activities in switchable antimicrobial and antifouling coatings. This action is particularly useful in biomedical coatings applications [170]. Herein, the functionalization of steel using the new matrix impacts significant antimicrobial activity against *S. epidermidis* and *E. coli* and also resistance to protein adsorption, bacterial adhesion, and microalgal attachment for instance *Amphora coffeaeformis*. Ngo and Grunlan in 2020 used a variety of strategies to introduce surface hydrophilization agents in their article titled Protein resistant polymeric biomaterials, including physisorption, hydrogel network development, surface grafting, layer-by-layer (LbL) assembly, and mixing base polymers with SMAs (surface modifying additives) [154]. Alex Cavallaro and coworkers [327] reviewed the common approaches for developing responsive and "smart" antibacterial surfaces, including photo-responsive, bio-responsive, temperature-responsive, pH-responsive surfaces, etc. [327] The application of stimuli-responsive polymers in infection treatment, drug delivery, and functional coatings have been explored [327]. And attempts are been made to improve adhesion, compatibility, and stability. Table 4 is a summary of the various technologies for the fabrication of antibacterial surfaces with their advantages and limitations.

Conclusion/Outlook

Observedly, the culture of routine environmental cleaning and sterilization measures has been linked with a reduction in surface contamination. However, adopting a well-documented cleaning and disinfecting procedure can still fail to achieve the required level of cleanliness, as organisms may sometimes out-maneuver the cleaning process, thus making the surfaces become re-contaminated immediately or soon after cleaning. For that reason, the fabrication of surfaces that can stop bacteria adhesion is confirmed to be the first line of defense against bacterial infection and is shown to be an effective way of avoiding bacterial infections and biofilm formation without the use of antibiotics. Over the years, many textured and chemically fabricated surfaces for antibiosis and antibacterial fouling have been achieved. Nonetheless, to adapt to various metals, alloys, polymers, ceramics, and environmental/operating conditions, new methods and advancements in antibacterial surface design are continuously in demand. This review has shown that present studies on antibacterial surfaces have developed remarkably and some of the techniques discussed are inspired by nature; animals and plants have fascinating strategies for preventing pathogens from colonizing their surfaces. Such bio-inspired approaches include the fabrication of micro/nanostructured surfaces to regulate the hydration layer, enabling them to be either superhydrophobic or superhydrophilic, mimicking the NO production in the endothelium, zwitterionic functionalities seen in the membrane, and the protein-repellant peptides.

These remarkable developments notwithstanding, there is still a need for improvement as highlighted in Section 5.0 and Table 4 where major limitations of some of the techniques are discussed. For instance, one major drawback is the durability of the antibacterial coatings; the coatings tend to fail after prolonged use. Currently, several approaches have been adopted to either retain or restore antifouling properties after failure including the introduction of self-healing [330,331] and smart cleaning [149] functionalities and the design of stimuli-responsive coatings which permits the regeneration of the antifouling performance by triggered release of foulants [332,333]. Further research needs to be done in terms of bacteria targeting, broad-spectrum, programmable, long-term stability, self-healing, environmentally friendly, and non-drug resistant techniques. In addition, efforts should be tailored towards practically possible methods which can be easily implemented on an industrial scale. It is important to note that in antibacterial surface fabrication, the surface characteristics of the target bacteria (such as bacteria structure, type, adhesion force, and featured proteins) are significant factors that will enable specific and selective action against biofilm formation.

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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

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