#### **Review**

Lihui Luo, Ying Zhou, Xiaoling Xu\*, Wanglin Shi\*, Jiahao Hu, Guo Li, Xi Qu, Yifan Guo, Xin Tian, Ahsan Zaman, David Hui, and Zuowan Zhou\*

# Progress in construction of bio-inspired physicoantimicrobial surfaces

https://doi.org/10.1515/ntrev-2020-0089 received September 17, 2020; accepted October 9, 2020

Abstract: Bacteria are widely distributed in the natural environment and the surfaces of objects, bringing about much trouble in our lives. Varies nanomaterials have been demonstrated good effect on killing microbe, but the consistency and stability seem to be improved. Recently, antibacterial effect on surfaces of some natural nanostructures was recognized, and more and more evidences were provided as a new type of bactericidal mechanism, the physical sterilization. The dragonfly and cicada wings have been found to possess the most exceptional antibacterial properties because of the specific nanostructure. Inspired by the biofunctions, researchers began to build a series of physico-antimicrobial surfaces on different materials to avoid the abuse of antibiotics and the environmental pollution of organic antibacterial agents. The physico-antimicrobial structure does not rely on chemical components, and a series of physicoantimicrobial models have been established. To deeply understand the physically bactericidal effect, this article

reviews a series of natural and biomimetic physical antibacterial surfaces and makes reasonable expectations for the application of such composite materials in constructing physical antibacterial surfaces.

Keywords: physico-antimicrobial, construction method, biomimetic structure

#### 1 Introduction

It is well known that bacteria can be found on the surface of all objects and often causes severe damage to human production and lives [1-3]. Currently, one of the most common antibacterial methods is using antibiotics. However, it is reported that bacterial antibiotic resistance is becoming more and more critical due to the excessive use of antibiotics in health care and agricultural activities. About 7,00,000 people die each year from antimicrobial resistance (AMR) infections, and it is estimated to be increased to 10 million by 2050 [4,5]. Furthermore, structural or spatially heterogeneous host population and the variability of antibiotic consumption will persist for longlasting coexistence [6]. Another prevalent method is to modify the surface of the material with the chemical antibacterial agent [7-11], which can bring a particular antibacterial effect to these materials. But the introduction of the inorganic or organic antibacterial agents may accompany with dissolution problems [12-14], such as the heavy metal ions or toxic chemical groups contained in those chemical antibacterial agent will be released from the surface of the material and inevitably pollute the environment [15]. Meanwhile, the addition of the antimicrobial agent as a second phase in the matrix material often lead to an increase in processing complexity and a decrease in particular properties [16–18].

On the basis of the aforementioned questions, we are looking forward to a new nonchemical, environmentally friendly [19], healthy [20], simple, and effective antibacterial technology, expecting that the surface of the material will have an excellent antibacterial effect based on physical

Lihui Luo, Ying Zhou, Jiahao Hu, Guo Li, Yifan Guo, Xin Tian, Ahsan Zaman: School of Materials Science and Engineering, Key Laboratory of Advanced Technologies of Materials (Ministry of Education), Southwest Jiaotong University, Chengdu 610031, China Xi Qu: China Academy of Space Technology, Beijing 100094, China David Hui: Department of Mechanical Engineering, University of New Orleans, New Orleans, LA 70148, United States of America

<sup>\*</sup> Corresponding author: Xiaoling Xu, School of Materials Science and Engineering, Key Laboratory of Advanced Technologies of Materials (Ministry of Education), Southwest Jiaotong University, Chengdu 610031, China, e-mail: xiaolingxu@switu.edu.cn

<sup>\*</sup> Corresponding author: Wanglin Shi, Shenzhou Space Biology Science & Technology Co., Ltd, Research Center of Beijing Space Engineering Technology, Beijing 100190, China, e-mail: shiwanglin@126.com

<sup>\*</sup> Corresponding author: Zuowan Zhou, School of Materials Science and Engineering, Key Laboratory of Advanced Technologies of Materials (Ministry of Education), Southwest Jiaotong University, Chengdu 610031, China, e-mail: zwzhou@swjtu.edu.cn, tel: +86 2887600454, fax: +86 2887600454

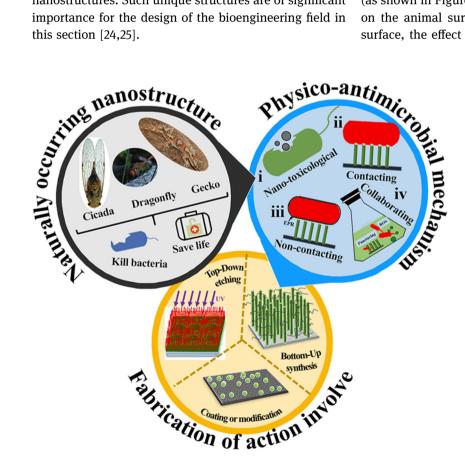
or even more simplistic mechanical action. This review article will explore various fabricated nanostructures surfaces and their underlying physical properties, aiding them to inhibit bacterial contamination. Bactericidal mechanisms and its effects on the physico-antimicrobial efficacy are also discussed. The three major aspects of physico-antimicrobial outlined in this review are shown in Figure 1.

# 2 Physico-antimicrobial material and its microfabrication and nanofabrication

As a proverb says, it is not the strongest of the species that survive, but the one most responsive to change. After several million years of evolution, the surface of many plants and animals were given some unique structures that make them magical to withstand external risks in harsh natural conditions [21–23]. A lot of research works pay great attention on natural and artificial antimicrobial nanostructures. Such unique structures are of significant importance for the design of the bioengineering field in this section [24,25].

The antibiofouling performance of these natural surfaces is attributed to their superhydrophobic and microtopographies or nanotopographies. The special properties aroused from the natural microstructures/nanostructures attracted many researchers and they began tostudy them [25]. Ma *et al.* [26] found that the surface of a taro leaf is a layer of polygonal epidermal cells. Different from lotus, such a unique uneven structure distribution makes the taro leaves much long lasting and prevent the adhesion of Gram-negative bacteria even in an entirely humid environment. Shark skin (The Sharklet AF<sup>TM</sup> PDMSe) [27–31] is also found to have antibacterial action toward the adherence of *Staphylococcus aureus*.

A series of work on natural bactericidal surfaces have been carried out since 2012. The cicada wings exhibiting nanoneedle array evenly distributed on its surface could make Gram-negative bacteria die in 5 min by direct physical contacting, first described by Mainwaring *et al.* [32]. The mechanobactericidal surfaces were successively observed on the surface of many other kinds of animals such as dragonfly wings, cicada wings, and gecko skin (as shown in Figure 2). Although the water contact angle on the animal surface is lower than that on the plant surface, the effect of antibacterial is not much different



**Figure 1:** Illustrative overview of the three major aspects of physico-antimicrobial outlined in this review. Note that the different components of this graphic are not drawn to scale.

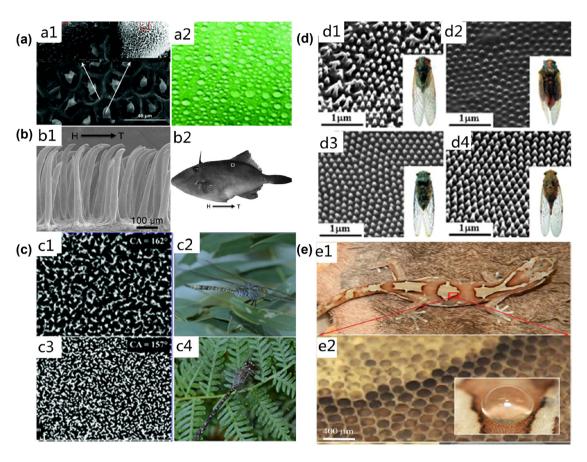


Figure 2: Summary of naturally occurring nanostructured antibiofouling surfaces: (a) taro leaf [26] and (b) *Navodon septentrionalis* filefish skin [44]; bactericidal surfaces: (c) different species of dragonfly wings [45,46]. (d) Different species of cicada wings [47], and (e) gecko foot with high adhesion [48].

[33], indicating that the antibacterial performance of natural antibacterial surfaces, especially the relationship between bactericidal performance and hydrophobicity, is not very close [34] (Figure 3).

# 2.1 Bio-inspired artificial nanostructures for physico-antimicrobial

Nature does always give plenty of inspiration [23]. Innovative studies from naturally occurring nanostructural surfaces over the past ten years have led to a next-generation bactericidal paradigm, which acts through contributing a series of the physico-antimicrobial surface [35,36].

#### 2.1.1 Top-down multiway etching

To replicate naturally occurring nanostructural behavior, researchers use inorganic, metallic, or polymeric materials

as the substrates to fabricate microsclaed or nanoscaled artificial structures [37,38]. Different arrangement densities on the surface of polymethyl methacrylate (PMMA) fabricate special nanostructures. Moreover, after quantitative analysis, in terms of PMMA, the optimal nanocolumn spacing is 130-380 nm. Likewise, the material having the antibacterial surface was placed in the bacterial suspension, and after 24 h, the bacterial content in the water was reduced by 50%. The Epstein group [39] bonded a layer of electron-patterned polyethylene glycol hydrogel film to the slide, which effectively inhibited the adhesion of bacteria to the surface. Chemical hydrothermal etching has been further explored by scientists. After being modified by solution-phase etching, Cu (used a soft H<sub>2</sub>O<sub>2</sub> and HCl solution) [40] and aluminum alloy (with a mixed solution of oxalic acid and then infiltrated it with the potassium permanganate solution) [41] substrates can be fabricated toward nanostructures of layered-triangular prism arrays with rough surfaces. The Cu substrate also displays selective superhydrophobic performance via facile solutionphase etching. Peng et al. [42] also etched a layered

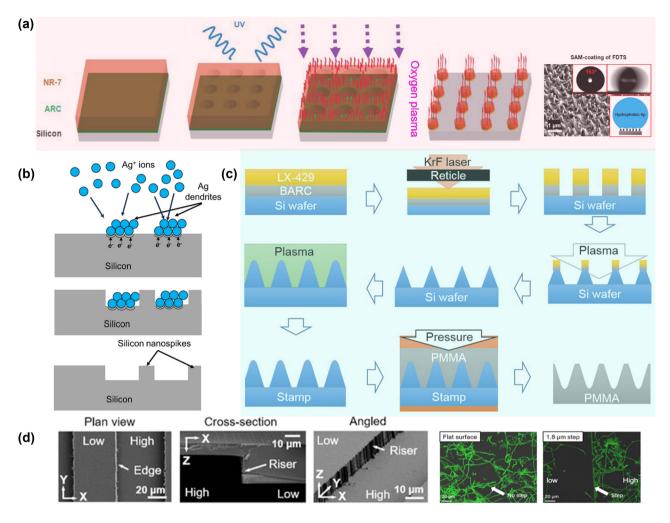


Figure 3: Schematic representation of the hysico-antimicrobial nanostructures that can be designed by top-down multiway etching. (a) The fabrication of monotonous polymer nanostructures on a planar surface [49]. (b) Schematics showing the two steps of Mac-Etch (metal-assisted chemical etching method) on silicon [50]. (c) The details of the nanopatterning process on the 8 in. Si wafer [51]. (d) Scanning electron microscope (SEM) images of a fabricated topographical step and images of the positions visited by the bacteria [52].

triangular prism array nanostructure using hot ammonia solution etching. Based on this, Arisoy  $et\ al.$  [43] printed the surface of shark skin with polymer and ceramic composites and added  $TiO_2$  nanoparticles. Compared to smooth films, the shark skin without nanoparticles reduced the adhesion of *Escherichia coli* by 70%. But  $TiO_2$  nanoparticles exposed to UV light for an hour will kill more than 95% of *E. coli* and 80% of *S. aureus*. The schematic representation of the top-down multiway etching is shown in Figure 3.

#### 2.1.2 Bottom-up chemical synthesis

Since the typical size of a single bacterium is about 1  $\mu$ m, most bacteria are nanoscale in a certain dimension

[53-55]. Therefore, when bacteria are in direct contact with many inorganic nanomaterials, their morphology and specific biological functions are significantly affected [56-58]. As an antibacterial material, the antibacterial mechanism of nano-ZnO has been widely concerned by researchers [59]. It can be considered that the small size effect of nanoparticles is the reason why ZnO has antibacterial effect in the absence of light. It has been found that the direct contact between ZnO nanoparticles and bacterial cell walls leads to cell injury. The antibacterial effects of ZnO nanoparticles with different morphologies are also different, for example, flower-like inactivation of E. coli and S. aureus is much better than spherical ZnO-NPS or rod-like ZnO-NPS. Carbon nanotubes [60-64], graphene [65,66], graphene oxide [22,67–69] (Figure 4), and the latest two-dimensional material MXenes [70]

have also reported to possess mechanical and physical antibacterial capabilities. Carbon nanotubes are related to nanodarts, and graphene is associated with nanoblades [71-73]. In 2007, Elimelech and coworkers [60] first demonstrated that single-walled carbon nanotubes (SWCNTs) destroy the cell membrane of *E. coli* by piercing and then excluding metal toxicity and oxidative stress. Scientists have ruled out experimental variables and possible factors, for example, the natural bacterial fading and material toxicity have been excluded. It was found that SWCNTs with a diameter of 8.3 Å have higher physical properties than direct contact with multiwalled carbon nanotubes (MWCNTs) [74,75]. The former was detected to have more leakage of genetic material after cell membrane destruction. Briefly speaking, the bactericidal ability of the SWCNTs is much better than that of the MWCNTs.

In addition to one-dimensional CNTs, the two-dimensional materials also have been found to display mechanically bactericidal effects. Among the two-dimensional materials, graphene oxides (GO) vertically covered on substrate have been widely investigated [66, 76–80]. For example, on the stainless steel substrate, the randomly oriented GO nanosheets are sharped like blades, which can cut through the cell membrane of bacteria, killing bacteria. Huang and coworkers found [67] that reduce graphene oxide (rGO) reduced by inductively coupled low temperature hydrogen plasma without using any other chemicals has significantly higher bactericidal activity, as depicted in Figure 4. This mechanism for the enhancing antibacterial activity was due to the decomposition of the

thin graphene oxide layer into smaller layers by plasma treatment. Because of these defects and small ridges formed on the surface, the bacteria suffer destructive membrane damage.

In fact, the role of physico-antimicrobial are still vaguely for the antibacterial effect of nanomaterials [81]. Most importantly, the potential influences on the synergistically antibacterial mechanisms are still unclearly. Conversely, the size dependence of multidimensional materials also makes it difficult to determine which part has the influence on the destruction of bacterial cell membrane accurately [82].

#### 2.1.3 Surface topography coatings and modification

There are many ways to grow nanostructures on the surface of different kind of substrates, including hydrothermal synthesis [83], chemical deposition, self-assembly, and inkjet printing and 3D printing. What is more, modification on those prepared nanostructures has also gradually emerged for fabricating composites [84–87], and the addition of various antibacterial agents can also improve its function [88,89].

The hydrothermal method can synthesize various nano-array structures such as nano-grass, nano-blade, and nano-pyramid on different substrates [90,91]. As a material with several morphological structures, ZnO has been found having a bactericidal effect on columnar needle-like or blade-like nano-array structures synthesized by hydrothermal processes [82,92,93]. Wu Shuilin's

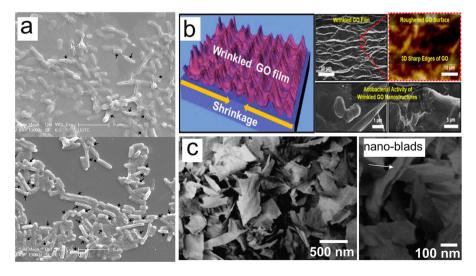


Figure 4: (a) SEM images of E. coli after incubating with GO and r-GO at 0.2 mg/mL for 4 h, respectively [67]. (b) The wrinkled surface of graphene films may directly affect bacterial viability by means of various interactions of bacterial cells with graphene sheets [68]. (c) Graphene nanoblades in the form of graphene nanowalls deposited on stainless steel substrates [66].

group [94] prepared a ZnO array structure with antibacterial and biocompatible properties by developing a seed layer on the titanium substrate by atomic deposition and then hydrothermally growing ZnO rods. Tetra-needle-like ZnO whisker (T-ZnO w) added prepared by gas pressure sintering process was reported to have good antibacterial properties. In the recent work in our group, a nanobladelike surface made of Zn-Al layers of double hydroxides was prepared on stainless steel sheet by chemical bath deposition (CBD) to mimic the nanoblade structure of the pitcher plant's neck [95]. The electric field-assisted hydrothermal growth method has been used to construct gradient morphologies such as hexagonal prisms and hexagonal pyramids on stainless steel sheets as illustrated in Figure 5. Although the density is similar  $(1.9 \times 10^9 - 2.4 \times 10^9 \,\text{rod cm}^2)$  [96], the bactericidal rate was greatly improved as the average tip width of the nanorods decreasing from 137 to 38 nm. It has been proved that there are mechanically bactericidal effect and self-cleaning effect in such structure.

Many other methods and techniques of microfabrication and nanofabrication that build these artificial nanostructures are proposed [97]. Wang research group [98] synthesized fluorinated multi-walled carbon nanotubes (MWCNTs-PFOL). By grafting perfluoro-*n*-alcohol to the surface of the MWCNTs, a mixture comprising polyurethane prepolymer, perfluoro-*n*-alcohol, ethylene glycol, acetone, and toluene was then spin coated onto the glass substrate together with MWCNTs-PFOL to give a corallike nanostructure. Zhao et al. [99] and Wong et al. [100] used the self-assembly technology and the jet assembly technique to construct unique nanostructures, respectively. The advantages of chemical deposition and assembly methods have lowered cost and made the mass production easy, but the resulting nanostructures are often not regular enough. Vafaei et al. [101] and Zhao et al. [102] used inkjet or 3D printing technology, which was a trendy material preparation technology in recent years, to print three-dimensional nanostructures on the substrate, respectively. It was easy to operate and accurate in preparation, but costly. Besides, it was difficult to obtain a nanoscaled structure. A porous polymer film with a uniform pore size array was prepared by the gas-phase method, utilizing the self-assembly behavior of the polymer and controlling droplet condensation. A uniform micron-sized porous structure surface was prepared, which limited the respiration of *Pseudomonas aeruginosa*, when the pore diameter was  $5-11 \,\mu\text{m}$ , thereby killing the bacteria [103].

In addition, there is a method combining these techniques. Specific steps include the following: (i) prepare a layer of easily etched material on the surface of the

substrate and (ii) etch the layer into a nanostructure. Zhang et al. [104] first coated a layer of fluorosilane on the surface of TiO2-SWNT and then irradiated the surface with ultraviolet light. After the fluorosilane was gradually decomposed, a particular structure at the nanometer level was obtained. Microwave plasma chemical vapor deposition (MPCVD) coupled with RIE had replicated the nanopattern of cicada wings on a diamond nanocone-patterned surfaces. The average structure heights were recorded to be 1.6 µm and the widths 350-750 nm [105]. To simulate the low surface energy of the natural pitcher plant neck, the nanoblades with (heptadecafluoro-1,1,2,2-tetrahydrodecyl)-trimethoxy silane were prepared [95], and with good self-cleaning performance, it could effectively remove bacterial debris and prevent biological adhesion.

## 3 Mechanisms for the physicoantimicrobial effect

Depending on the type of species of naturally occurring nanostructured antibacterial surfaces, the combinatorial effects of antimicrobial activity manifest themselves as the total destruction of the cellular envelope (i.e., biocidal effect) or inhibition of growth (i.e., antibiofouling effect).

Overall, the killing mechanism of the physicomechanical antibacterial material or surface is seemly a combination of several factors including (i) microparticle's and nanoparticle's internalization or insertion, resulting in membrane dysfunction (i.e., nanotoxicological effects), (ii) contacting physical puncturing (also known as mechanobiocidal mechanism) (iii) noncontacting physical tearing, and (iv) collaborating with some conventional chemical destructive extraction includes damaging of oxidative stress, and so on. Since the antibacterial spectrum of this surficial or structural antibacterial has not been established, the interpretation of its antibacterial mechanism is also different. Here, we summarize research efforts in the proposed mechanism of the physico-antimicrobial material.

In terms of mechanical antibacterial of nanomaterial, nanoparticles affect the adhesion of cells to substrates (such as ECM, cells, or tissues) [106–108], the integrity of the internal skeleton of cells [109,110], and the ability of cells to migrate [71,111]. Liu et al. used a 2 nm atomic force microscope (AFM) to simulate a single collision event between cells and SWCNTs and found that a single

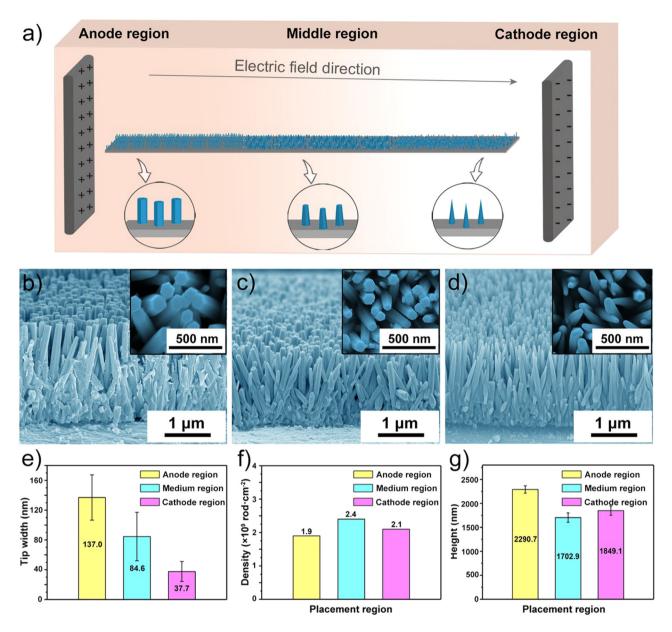


Figure 5: The experimental setup (a) and changes in ZnO NAs topography under the influence of setting position of samples. SEM images of (b) anode region, (c) middle region, and (d) cathode region; topographic parameters: (e) tip-width, (f) density, and (g) height, as determined by SEM [96].

collision between CNTs and cells was not sufficient to induce fatal damage [65]. Therefore, the author interprets the process of mechanically piercing bacterial cells by carbon nanotubes as a cumulative process of repeated punctures [53]. Simulation of individual interactions of CNTs with cellular lipid bilayer membranes generally agrees that CNTs will first enter the cell through either end of the tube structure, possibly by entering the cell in an approximately vertical manner [107,112–115]. For example, Luan *et al.* [116] used the simulation of molecular dynamics simulation, and the interaction process

between flake graphene and biological cells is revealed from the molecular level. In the main life activity of cells synthesizing secreted proteins, graphene sheets organize the hydrophobins to contact each other to recognize, thereby destroying cell metabolism and causing apoptosis. Figure 6a shows a schematic view showing a process of breaking a cell membrane by microparticle and nanoparticle insertion.

The contacting physical puncturing mechanical are proposed by the Ivanovo's group [117]. They used the flap surface as a model, assuming that all the nanopillars on

the arrays are evenly distributed and of the same height, since the bacterial cell size is generally 500-1,000 nm and the gap distance between the nanocolumn arrays is about 170 nm. Therefore, when the bacterial cells are in contact with the nanocolumn array, the cells cannot be in the gap. Because the height of the nanocolumn is about 200 nm, which is an order of magnitude higher than that of the cell membrane that is less than 10 nm thick, the bacterial cells cannot approach and contact the nanocolumn. It is impossible for the arrays to contact the substrate over a large area by a slight deformation of the cell membrane, lying on the substrate, and it is only possible to be placed in the air by the array of nanopillars, as shown in Figure 6b. Owning to its own weight and adsorption, when the bacteria approach the surface of the array, cell membrane begins to deform and stretch, and the part between the connected nanopillars begins to tear [118]. Once the cell membrane breaks, cell death finally occurs. Saccharomyces cerevisiae, a eukaryotic microorganism, can be ruptured by nanostructure geometry and has adhesion dependence. The process of interaction between cells and nanostructures is very similar to the physico-antimicrobial mechanism mentioned earlier [119].

The mechanical antibacterial model explained why the flap structure kills the Gram-negative bacteria, but Gram-positive bacteria show resistance. Cheeseman *et al.* [120] used giant unilamellar vesicles (GUVs) as a general

model of cell biology and also observed the phenomenon that the cells were torn by the nanostructures on the surface of the two wings, which further confirmed the principle of the contact mechanical antibacterial effect.

However, other recent studies [61,121] believed that bacteria will undergo a stress response when exposed to external mechanical stress, releasing a polysaccharidelike extracellular polymeric substance (EPS). It is believed that the bacterial membrane damage is caused by the strong adhesion between the nanocolumn and bacterial EPS, which is caused by the increased shear force when the bacteria attempt to move on the surface of the nanocolumn rather than the nanoarrays and bacterial cells directly pierced by contact (as shown in Figure 6c). Advanced electron microscopy, such as the application of TEM, makes this antibacterial process more intuitive. This noncontact physical antibacterial interpretation that relies on bacterial adhesion is also flawed. E. coli ruptured three to five minutes after its contact with the etched silicon array, but traced by concanavalin labeling revealed that bacteria did not have time to secrete EPS at this time [81]. In the latest research, Xie et al. [122] further proved that the arrays rely on puncture to achieve a rapid sterilization effect, but the sterilization speed of these uneven arrays are significantly faster than that of these ordered arrays. They believe that the bacteria need to go through three stages from falling on the ordered arrays surface to being killed, which takes a

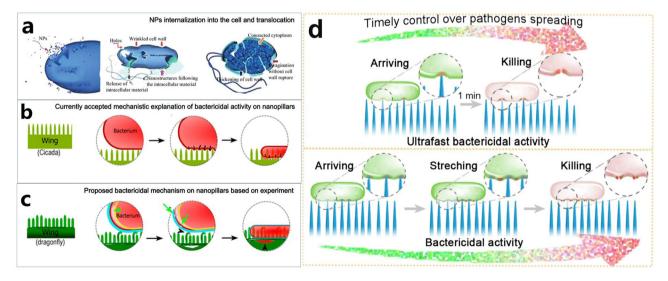


Figure 6: The proposed mechanism of physico-antimicrobial material activity. (a) Microparticle's and nanoparticle's internalization or insertion [116]. (b) The mechanism of bactericidal activity based on current accepted mechanistic models (also known as mechanobiocidal mechanism) using the cicada wing structure. (c) The proposed mechanism based on shearing of the membrane as the cell attempts to move across the nanotextured surface, while it is immobilized (also known as noncontacting physical tearing mechanism) [121]. (d) The proposed ultrafast bactericidal mechanisms of ZnO NAs with ordered and uneven topography [122].

long time. However, it only takes two stages to kill the bacteria on the uneven arrays surface with a short time. As a result, it shows ultrafast sterilization performance.

In fact, the vast differences in structural dimensions and contact angles between different materials indicate that no particular surface mode has a universal antibacterial effect on all types of microorganisms, so the mechanism of physico-antimicrobial is not totally illuminated and still controversial. But no matter what kind of physical antibacterial mechanism, we can easily find that the structure determines the performance. Without the particular structure of the surface of these materials. the physico-antimicrobial surface is bound to be challenging to form. In addition, there may be chemical reactions in the physical antibacterial process, but its role in the rapid sterilization process is very small. This is mainly because the process of chemical mechanism takes a long time, while the process of physical mechanism takes very short time. Before the chemical mechanism works, the bacteria are already killed.

# 4 Effects on the physicoantimicrobial efficacy

Although researchers have extensively analyzed and designed the biomimetic physico-antimicrobial effect of materials, most of the research works still focus on the composition and the structure of materials. We present here from these two aspects.

#### 4.1 Physicochemical effects

Silicon is widely used in semiconductor manufacturing processes, but there will be many spikes and edges during processing. However, these obstacles can be used in physico-antimicrobial fabrication. The surface of the ordinary untreated silicon substrate is not antibacterial [123,124]. After discovering that the unique structure of the biological surface has a bactericidal effect, many scientists etched on black silicon substrates to confirm biomimetic sterilization [32,46,125,126]. The nanostructure of the black silicon (bSi) surface fabricates with RIE that also reacted with red blood cells physically [81]. The nano-array spontaneous stress induced cell deformation and rupture without external chemical or mechanical intervention, which only took 3 min [127].

Because of its high biocompatibility, titanium is more used in the field of biological antibacterial [128]. Similar to the silicon substrate, the structure functionalization of titanium metal is mostly completed by etching [129–131]. But some materials build other structures on the titanium substrate, such as ZnO, to achieve a composite physicoantimicrobial efficacy [132]. The results of in vitro and in vivo experiments have confirmed that titanium substrates grown with ZnO nanoarrays have selective puncture of bacterial membranes ability. But these materials hardly cause harm to the human body. On the one hand, most of them are not currently used in the human body. On the other hand, researchers basically use biocompatible materials when constructing physico-antimicrobial structures, which further reduce the possibility of harm to the human body.

### 4.2 Topographical influence on the physicoantimicrobial efficacy

The neatly arranged array of cicada wings can only kill Gram-negative bacteria effectively [35], but not Grampositive bacteria. But the uneven array of dragonfly wings can kill both Gram-negative and Gram-positive bacteria. The difference of surface topography has a significant effect on the physico-antimicrobial activity.

By adjusting the etching time, etching pressure, and even the type of reactor, sundry geometry and morphology nanopillars array of black silicon spikes can be achieved, resulting in different physico-antimicrobial effects. Recently, different length-to-diameter array structures can be obtained in the black silicon surface by different plasma etching time [123,124]. 15, 30 and 45 min. An increase in etching time directly leads to silicon wafer surface nanostructures of varying heights (approximately 212, 475, and 610 nm), cap widths (about 21, 93, and 114 nm), and spacing (50, 107, and 140 nm, respectively) [81]. The experiment proved that the etching time obtained 85%, 85%, and 89% antibacterial rates, respectively. We also found in the work that although the prepared ZnO nanoarrays have the same density, different morphologies of single nanorods have an unusual ability to destroy bacteria. That is, the integrity of the cell membrane on the hexagonal prism-shaped nanorods has not been extremely damaged, and the number of bacterial debris on the surface of prismoid nanoarrays (prismoid NAs) has increased significantly. E. coli cells on the surface of pyramid nanoarrays (Pyramid NAs) were severely damaged, the cells were dried up, and multiple sites of the cell membrane were "penetrated" by the pyramid NAs, resulting in the flow of cell contents and the cell death. There is a great difference in the sterilization ability between uneven arrays and ordered arrays [122]. Besides, the wrinkle structure of the GO surface has also been proved to be a "trap" [68], which has a bactericidal effect. Different roughness has different intensity of action on different bacteria. 500 nm roughness level is sterilized for E. coli and S. aureus. The result is better, and the roughness of 845 nm is better for the bactericidal effect of Mycobacterium smegmatis.

## 5 Outlook and perspectives

With the increasing demand of health and the human life, the use of antibacterial and bactericidal materials is increasing. It is hoped that an excellent antibacterial effect can be achieved by the construction of a unique physical structure on the surface of the material [34]. Inspired by a variety of antibiofouling and mechanobactericidal micronano structures on the surface of natural plants and animals, scientists have also constructed a series of physical antibacterial surfaces and proposed different physical antibacterial mechanisms and models. These physico-antimicrobial surfaces are structurally characterized by a distribution of arrays of columnar or sharp blades. However, in terms of self-cleaning recycling, in addition to photocatalysis, more self-cleaning technologies need to be developed. Moreover, the broad spectrum of the physico-antimicrobial effect needs further verification, and the structure-activity relationship is not comprehensive enough. Therefore, it will be a gigantic challenge to prepare a multifunctional composite material based on a rapid physico-antimicrobial structure. But once this material can be prepared, its market prospects will be very enormous.

Acknowledgements: This research was financially supported by the National Natural Science Foundation of China (No. 51772251), the Science and Technology Planning Project of Sichuan Province (Nos. 2020ZDZX0008 and 2018GZ0431), the Pre-research Foundation of Manned Spaceflight (No. 020301), the Space Station Engineering Aerospace Technology Test Field Project and the Fundamental Research Funds for the Central Universities (Nos. 2019XJ02, 2682020XG05).

Conflict of interest: The authors declare no conflict of interest regarding the publication of this paper.

#### References

- Bazaka K, Jacob MV, Crawford RJ, Ivanova EP. Efficient surface modification of biomaterial to prevent biofilm formation and the attachment of microorganisms. Appl Microbiol Biotechnol. 2012:95(2):299-311.
- Arciola CR, Campoccia D, Speziale P, Montanaro L, Costerton JW. Biofilm formation in Staphylococcus implant infections. A review of molecular mechanisms and implications for biofilm-resistant materials. Biomaterials. 2012:33(26):5967-82.
- [3] Arciola CR. Campoccia D. Montanaro L. Implant infections: Adhesion, biofilm formation and immune evasion. Nat Rev Microbiol. 2018;16(7):397-409.
- Sugden R, Kelly R, Davies S. Combatting antimicrobial resistance globally. Nat Microbiol. 2016;1(10):16187.
- [5] Duval RE, Grare M, Demore B. Fight against antimicrobial resistance: We always need new antibacterials but for right bacteria. Molecules. 2019;24(17):3152.
- Krieger MS, Denison CE, Anderson TL, Nowak MA, Hill AL. Population structure across scales facilitates coexistence and spatial heterogeneity of antibiotic-resistant infections. bioRxiv. 2020;16(7):469171.
- [7] Chibeu A, Agius L, Gao A, Sabour PM, Kropinski AM, Balamurugan S. Efficacy of bacteriophage LISTEX (TM) P100 combined with chemical antimicrobials in reducing Listeria monocytogenes in cooked turkey and roast beef. Int J Food Microbiol. 2013;167(2):208-14.
- [8] Smarr MM, Sundaram R, Honda M, Kannan K, Louis GMB. Urinary concentrations of parabens and other antimicrobial chemicals and their association with couples' fecundity. Environ Health Perspect. 2017;125(4):730-6.
- Wright MH, Fetzer C, Sieber SA. Chemical probes unravel an antimicrobial defense response triggered by binding of the human opioid dynorphin to a bacterial sensor kinase. J Am Chem Soc. 2017;139(17):6152-9.
- Behbahani BA, Yazdi FT, Vasiee A, Mortazavi SA. Oliveria decumbens essential oil: Chemical compositions and antimicrobial activity against the growth of some clinical and standard strains causing infection. Microb Pathogen. 2018;114:449-52.
- Zhao L, Chu PK, Zhang Y, Wu Z. Antibacterial coatings on [11] titanium implants. J Biomed Mater Res Part B Appl Biomater. 2009;91B(1):470-80.
- Yang H, Liu C, Yang D, Zhang H, Xi Z. Comparative study of cytotoxicity, oxidative stress and genotoxicity induced by four typical nanomaterials: The role of particle size, shape and composition. J Appl Toxicol. 2009;29(1):69-78.
- Xia T, Kovochich M, Liong M, Maedler L, Gilbert B, Shi H, et al. Comparison of the mechanism of toxicity of zinc oxide and cerium oxide nanoparticles based on dissolution and oxidative stress properties. ACS Nano. 2008;2(10):2121-34.
- Wu B, Wang Y, Lee Y-H, Horst A, Wang Z, Chen D-R, et al. [14] Comparative Eco-toxicities of nano-ZnO particles under aquatic and aerosol exposure modes. Environ Sci Technol. 2010;44(4):1484-9.
- Zhang D, Chen L, Zang C, Chen Y, Lin H. Antibacterial cotton fabric grafted with silver nanoparticles and its excellent

- laundering durability. Carbohydr Polym. 2013;92(2):2088-94.
- [16] Inkinen J, Makinen R, Keinanen-Toivola MM, Nordstrom K, Ahonen M. Copper as an antibacterial material in different facilities. Lett Appl Microbiol. 2017;64(1):19-26.
- [17] Liu X, Xia W, Jiang Q, Yu P, Yue L. Chitosan oligosaccharide-Nchlorokojic acid mannich base polymer as a potential antibacterial material. Carbohydr Polym. 2018;182:225-34.
- [18] Shi J, Yang D, Jiang Z, Jiang Y, Liang Y, Zhu Y, et al. Simultaneous size control and surface functionalization of titania nanoparticles through bioadhesion-assisted bio-inspired mineralization. J Nanopart Res. 2012;14(9):1-13.
- Hasan J, Crawford RJ, Lvanova EP. Antibacterial surfaces: The quest for a new generation of biomaterials. Trends Biotechnol. 2013;31(5):31-40.
- Pantic S, Skodric SR, Loncar Z. Pantic I. Neurotoxocity, nephrotoxicity, and hepatotoxicity of copper-based nanoparticles: Potential implications in molecular medicine and neurosciences. Rev Adv Mater Sci. 2019;58(1):201-5.
- Wei X, Huang T, Nie J, Yang J-h, Qi X-d, Zhou Z-w, et al. Bioinspired functionalization of microcrystalline cellulose aerogel with high adsorption performance toward dyes. Carbohydr Polym. 2018;198:546-55.
- [22] Ceylan H, Kocabey S, Tekinay AB, Guler MO. Surface-adhesive and osteogenic self-assembled peptide nanofibers for bioinspired functionalization of titanium surfaces. Soft Matter. 2012;8(14):3929-37.
- Lee SJ, Ha N, Kim H. Superhydrophilic-superhydrophobic [23] water harvester inspired by wetting property of cactus stem. ACS Sustainable Chem Eng. 2019;7(12):10561.
- Nagel JKS, Rose C, Beverly C, Pidaparti R. Bio-inspired design pedagogy in engineering. In: Schaefer D, Coates G, Eckert C, editors. Design education today: technical contexts, programs and best practices. Cham: Springer International Publishing; 2019. p. 149-78.
- [25] Shang L, Zhang W, Xu K, Zhao Y. Bio-inspired intelligent structural color materials. Mater Horizons. 2019:6(5):945-58.
- Ma J, Sun Y, Gleichauf K, Lou J, Li Q. Nanostructure on taro leaves resists fouling by colloids and bacteria under submerged conditions. Langmuir. 2011;27(16):10035-40.
- [27] Chung KK, Schumacher JF, Sampson EM, Burne RA, Antonelli PJ, Brennana AB. Impact of engineered surface microtopography on biofilm formation of Staphylococcus aureus. Biointerphases. 2007;2(2):89-94.
- Darmanin T, Guittard F. Superhydrophobic and superoleophobic properties in nature. Mater Today. 2015;18(5):273-85.
- [29] Wen L, Weaver JC, Lauder GV. Biomimetic shark skin: Design, fabrication and hydrodynamic function. J Exp Biol. 2014;217(10):1656-66.
- Lee M. Shark skin: Taking a bite out of bacteria. In: Lee M, editor. Remarkable Natural Material Surfaces and Their Engineering Potential. Cham: Springer International Publishing; 2014. p. 15-27.
- [31] Chien H-W, Chen X-Y, Tsai W-P, Lee M. Inhibition of biofilm formation by rough shark skin-patterned surfaces. Colloids Surf B-Biointerfaces. 2020;186:110738.
- Mainwaring DE, Song HaN, Webb H, Jakubov T, Tobin M, Lamb RN, et al. The nature of inherent bactericidal activity:

- Insights from the nanotopology of three species of dragonfly. Nanoscale. 2016;8(12):6527-34.
- [33] Dalawai SP, Aly MAS, Latthe SS, Xing R, Sutar RS, Nagappan S, et al. Recent Advances in durability of superhydrophobic self-cleaning technology: A critical review. Prog Org Coat. 2020;138:105381.
- Jaggessar A, Shahali H, Mathew A, Yarlagadda PKDV. Biomimicking nano and micro-structured surface fabrication for antibacterial properties in medical implants. J Nanobiotechnol. 2017;15(1):34.
- Tripathy A, Sen P, Su B, Briscoe WH. Natural and bioinspired [35] nanostructured bactericidal surfaces. Adv Colloid Interface Sci. 2017;248:85-104.
- [36] Lau D, Jian W, Yu Z, Hui D. Nano-engineering of construction materials using molecular dynamics simulations: Prospects and challenges. Composites Part B. 2018;143:282-91.
- [37] Cheung H-Y, Lau K-T, Lu T-P, Hui D. A critical review on polymer-based bio-engineered materials for scaffold development. Composites Part B. 2007;38(3):291-300.
- [38] Jia F, Li G, Yang B, Yu B, Shen Y, Cong H. Investigation of rare earth upconversion fluorescent nanoparticles in biomedical field. Nanotechnol Rev. 2019;8(1):1-17.
- [39] Epstein AK, Hochbaum AI, Kim P, Aizenberg J. Control of bacterial biofilm growth on surfaces by nanostructural mechanics and geometry. Nanotechnology. 2011;22(49):494007.
- [40] Liu L, Xu F, Ma L. Facile fabrication of a superhydrophobic Cu Surface via a selective etching of high-energy facets. J Phys Chem C. 2012;116(35):18722-7.
- [41] Huang Z, Sun L, Luo Y, Liu Y, Wu R, Yuan Z. Preparation of aluminum alloy super-hydrophobic surface film by a acid etching method. J Hunan Univ Technol. 2011;25(4):5-8.
- [42] Peng S, Deng W. A simple method to prepare superamphiphobic aluminum surface with excellent stability. Colloids Surf A. 2015;481:143-50.
- Arisoy FD, Kolewe KW, Homyak B, Kurtz IS, Schiffman JD, Watkins JJ. Bioinspired photocatalytic shark-skin surfaces with antibacterial and antifouling activity via nanoimprint lithography. ACS Appl Mater Interfaces. 2018;10(23):20055-63.
- [44] Cai Y, Lin L, Xue Z, Liu M, Wang S, Jiang L. Filefish-inspired surface design for anisotropic underwater oleophobicity. Adv Funct Mater. 2014;24(6):809-16.
- [45] Vi Khanh T, Geeganagamage NM, Baulin VA, Vongsvivut J, Tobin MJ, Luque P, et al. The susceptibility of Staphylococcus aureus CIP 65.8 and pseudomonas aeruginosa ATCC 9721 cells to the bactericidal action of nanostructured calopteryx haemorrhoidalis damselfly wing surfaces. Appl Microbiol Biotechnol. 2017;101(11):4683-90.
- [46] Elbourne A, Crawford RJ, Ivanova EP. Nano-structured antimicrobial surfaces: From nature to synthetic analogues. J Colloid Interface Sci. 2017;508:603-16.
- [47] Sun M, Watson GS, Zheng Y, Watson JA, Liang A. Wetting properties on nanostructured surfaces of cicada wings. J Exp Biol. 2009;212(19):3148-55.
- [48] Watson GS, Cribb BW, Schwarzkopf L, Watson JA. Contaminant adhesion (aerial/ground biofouling) on the skin of a gecko. J R Soc Interface. 2015;12:108.

- Du K, Jiang Y, Liu Y, Wathuthanthri I, Choi C-H. Manipulation of the superhydrophobicity of plasma-etched polymer nanostructures. Micromachines. 2018;9(6):304.
- [50] Hu H, Siu VS, Gifford SM, Kim S, Lu M, Meyer P, et al. Bioinspired silicon nanospikes fabricated by metal-assisted chemical etching for antibacterial surfaces. Appl Phys Lett. 2017;111(25):253701.
- Kim S, Jung UT, Kim S-K, Lee J-H, Choi HS, Kim C-S, et al. [51] Nanostructured multifunctional surface with antireflective and antimicrobial characteristics. ACS Appl Mater Interfaces. 2015;7(1):326-31.
- Chang Y-R, Weeks ER, Barton D, Dobnikar J, Ducker W. Effect of topographical steps on the surface motility of the bacterium pseudomonas aeruginosa. ACS Biomater Sci Eng. 2019;5(12):6436-45.
- [53] Zhu Y, Ramasamy M, Yi DK. Antibacterial activity of ordered gold nanorod arrays. ACS Appl Mater Interfaces. 2014;6(17):15078-85.
- Talaro KP. Foundations in microbiology. Mc-Graw-Hill: Higher Education Press; 2013.
- Shuster J, Johnston CW, Magarvey NA, Gordon RA, Barron K, [55] Banerjee NR, et al. Structural and chemical characterization of placer gold grains: Implications for bacterial contributions to grain formation. Geomicrobiol J. 2015;32(2):158-69.
- [56] Li W, Zhao G, Zhang Q. Research progress and prospect on application of nanometer antibacterial materials in domestic woodworking industry. Mater Rev. 2015;29(10A):11-7.
- Zhou X-X, Liu R, Liu J-F. Rapid chromatographic separation of [57] dissoluble Ag(I) and silver-containing nanoparticles of 1-100 nanometer in antibacterial products and environmental waters. Environ Sci Technol. 2014;48(24):14516-24.
- Wei L, Chen X, Gao X, Guo R, Xu B. Preparation of Ag/SiO<sub>2</sub> powder with light color and antibacterial performance. Powder Technol. 2014;253:424-8.
- [59] Pantic S, Skodric SR, Loncar Z, Pantic I. Zinc oxide nanoparticles: Potential novel applications in cellular physiology, pathology, neurosciences and cancer research. Rev Adv Mater Sci. 2019;58(1):17-21.
- [60] Kang S, Pinault M, Pfefferle LD, Elimelech M. Single-walled carbon nanotubes exhibit strong antimicrobial activity. Langmuir. 2007;23(17):8670-3.
- [61] Kang S, Herzberg M, Rodrigues DF, Elimelech M. Antibacterial effects of carbon nanotubes: Size does matter. Langmuir. 2008;24(13):6409-13.
- [62] Kang S, Mauter MS, Elimelech M. Physicochemical determinants of multiwalled carbon nanotube bacterial cytotoxicity. Environ Sci Technol. 2008;42(19):7528-34.
- [63] Chen H, Wang B, Gao D, Guan M, Zheng L, Ouyang H, et al. Broad-spectrum antibacterial activity of carbon nanotubes to human gut bacteria. Small. 2013;9(16):2735-46.
- Lau KT, Chipara M, Ling HY, Hui D. On the effective elastic moduli of carbon nanotubes for nanocomposite structures. Composites Part B. 2004;35(2):95-101.
- Liu S, Wei L, Hao L, Fang N, Chang MW, Xu R, et al. Sharper [65] and faster "Nano Darts" kill more bacteria: A study of antibacterial activity of individually dispersed pristine singlewalled carbon nanotube. ACS Nano. 2009;3(12):3891-902.
- Akhavan O, Ghaderi E. Toxicity of graphene and graphene oxide nanowalls against bacteria. ACS Nano. 2010;4(10):5731-6.

- [67] Ke Z, Ma Y, Zhu Z, Zhao H, Wang Q, Huang Q. Non-thermal hydrogen plasma processing effectively increases the antibacterial activity of graphene oxide. Appl Phys Lett. 2018;112(1):013701.
- [68] Zou F, Zhou H, Jeong DY, Kwon J, Eom SU, Park TJ, et al. Wrinkled Surface-Mediated Antibacterial Activity of Graphene Oxide Nanosheets. ACS Appl Mater Interfaces. 2017:9(2):1343-51.
- Kaneda M, Lu X, Cheng W, Zhou X, Bernstein R, Zhang W, et al. Photografting graphene oxide to inert membrane materials to impart antibacterial activity. Environ Sci Technol Lett. 2019;6(3):141-7.
- [70] Rasool K, Helal M, Ali A, Ren CE, Gogotsi Y, Mahmoud KA. Antibacterial activity of Ti<sub>3</sub>C<sub>2</sub>T<sub>x</sub> MXene. ACS Nano. 2016;10(3):3674-84.
- Lin N, Berton P, Moraes C, Rogers RD, Tufenkji N. Nanodarts, [71] nanoblades, and nanospikes: Mechano-bactericidal nanostructures and where to find them. Adv Colloid Interface Sci. 2018;252:55-68.
- Meng F, Wei W, Chen J, Chen X, Xu X, Jiang M, et al. Growth of Fe<sub>3</sub>O<sub>4</sub> nanosheet arrays on graphene by a mussel-inspired polydopamine adhesive for remarkable enhancement in electromagnetic absorptions. RSC Adv. 2015;5(122):101121-6.
- [73] Lau KT, Hui D. Effectiveness of using carbon nanotubes as nano-reinforcements for advanced composite structures. Carbon. 2002;40(9):1605-6.
- Lau AK-T, Hui D. The revolutionary creation of new advanced [74] materials - carbon nanotube composites. Composites, Part B. 2002;33(4):263-77.
- [75] Zhao W, Zhu L, Lu Y, Zhang L, Schuster RH, Wang W. Magnetic nanoparticles decorated multi-walled carbon nanotubes by bio-inspired poly(dopamine) surface functionalization. Synth Met. 2013;169:59-63.
- [76] Yan Y, Li C, Wu H, Du J, Feng J, Zhang J, et al. Montmorillonitemodified reduced graphene oxide stabilizes copper nanoparticles and enhances bacterial adsorption and antibacterial activity. ACS Appl Bio Mater. 2019;2(5): 1842-9.
- [77] Guo Y, Peng F, Wang H, Huang F, Meng F, Hui D, et al. Intercalation polymerization approach for preparing graphene/polymer composites. Polymers. 2018;10(1):61.
- [78] Mohan VB, Lau K-t, Hui D, Bhattacharyya D. Graphene-based materials and their composites: A review on production, applications and product limitations. Composites Part B. 2018;142:200-20.
- [79] Ahmad H, Fan M, Hui D. Graphene oxide incorporated functional materials: A review. Composites Part B. 2018:145:270-80.
- [80] Kim H, Jang Y, Lee DY, Moon JH, Choi JG, Spinks GM, et al. Bio-inspired stretchable and contractible tough fiber by the hybridization of GO/MWNT/polyurethane. ACS Appl Mater Interfaces. 2019;11(34):31162-8.
- Linklater DP, Juodkazis S, Rubanov S, Ivanova EP. Comment [81] on "Bactericidal effects of natural nanotopography of dragonfly wing on Escherichia coli". ACS Appl Mater Interfaces. 2017;9(35):29387-93.
- [82] Ma J, Dong Y, Bao Y, Zhao Y, Liu C. Tunable microstructure of polyacrylate/ZnO nanorods composite emulsion and its filmforming properties. Prog Org Coat. 2019;135:382-91.

- [83] Ghouri Z, Motlak M, Afaq S, Barakat N, Abdala A. Templatefree synthesis of Se-nanorods-rGO nanocomposite for application in supercapacitors. Nanotechnol Rev. 2019;8:661-70.
- [84] Zhang S, Kai C, Liu B, Zhang S, Wei W, Xu X, et al. Preparation, characterization and antibacterial properties of cellulose membrane containing N-halamine. Cellulose. 2019;26(9):5621-33.
- Xie X, Gou G, Wei X, Zhou Z, Jiang M, Xu X, et al. Influence of pretreatment of rice straw on hydration of straw fiber filled cement based composites. Constr Build Mater. 2016;113:449-55.
- Wang X, Jiang M, Zhou Z, Gou J, Hui D. 3D printing of polymer matrix composites: A review and prospective. Composites Part B. 2017;110:442-58.
- Ngo TD, Kashani A, Imbalzano G, Nguyen KTQ, Hui D. Additive manufacturing (3D printing): A review of materials, methods, applications and challenges. Composites Part B. 2018:143:172-96.
- Hou Y-X, Abdullah H, Kuo D-H, Leu S-J, Gultom NS, Su C-H. [88] A comparison study of SiO<sub>2</sub>/nano metal oxide composite sphere for antibacterial application. Composites Part B. 2018:133:166-76.
- [89] Hong D, Cao G, Qu J, Deng Y, Tang J. Antibacterial activity of Cu<sub>2</sub>O and Ag co-modified rice grains-like ZnO nanocomposites. J Mater Sci Technol. 2018;34(12):2359-67.
- [90] Zhu S, Zhao J, Zhang X, Cui H, Zhou Z, Controllable synthesis of ZnO nanorod-nanosheets hierarchical nanostructure on a stainless steel mesh via a facile hydrothermal growth. MATEC Web of Conferences. 2016;67:02007.
- Ossai CI, Raghavan N. Nanostructure and nanomaterial characterization, growth mechanisms, and applications. Nanotechnol Rev. 2018;7(2):209-31.
- Yu Q, Xu X, Wang C, Ma Y, Hui D, Zhou Z. Remarkably improvement in antibacterial activity by synergistic effect in n-Cu@T-ZnO nanocomposites. Composites Part B. 2017;110:32-8.
- Krishnamurthi P, Raju Y, Khambhaty Y, Manoharan PT. Zinc oxide-supported copper clusters with high biocidal efficacy for Escherichia coli and Bacillus cereus. ACS Omega. 2017:2(6):2524-35.
- Li J, Tan L, Liu X, Cui Z, Yang X, Yeung KWK, et al. Balancing bacteria-osteoblast competition through selective physical puncture and biofunctionalization of ZnO/polydopamine/ arginine-glycine-aspartic acid-cysteine nanorods. ACS Nano. 2017;11(11):11250-63.
- Xie Y, Li J, Bu D, Xie X, He X, Wang L, et al. Nepenthes-inspired [95] multifunctional nanoblades with mechanical bactericidal, self-cleaning and insect anti-adhesive characteristics. RSC Adv. 2019;9(48):27904-10.
- Yuan X, Wei W, Fanbin M, Xi Q, Jinyang L, Li W, et al. Electricfield assisted growth and mechanical bactericidal performance of ZnO nanoarrays with gradient morphologies. Nanotechnol Rev. 2019;8(1):315-26.
- [97] Behdinan K, Moradi-Dastjerdi R, Safaei B, Qin Z, Chu F, Hui D. Graphene and CNT impact on heat transfer response of nanocomposite cylinders. Nanotechnol Rev. 2020;9(1):41-52.
- Wang X, Hu H, Ye Q, Gao T, Zhou F, Xue Q. Superamphiphobic coatings with coralline-like structure enabled by one-step

- spray of polyurethane/carbon nanotube composites. J Mater Chem. 2012;22(19):9624-31.
- [99] Zhao J, Ma L, Millians W, Wu T, Ming W. Dual-functional antifogging/antimicrobial polymer coating. ACS Appl Mater Interfaces. 2016;8(13):8737-42.
- [100] Wong WSY, Stachurski ZH, Nisbet DR, Tricoli A. Ultra-durable and transparent self-cleaning surfaces by large-scale selfassembly of hierarchical interpenetrated polymer networks. ACS Appl Mater Interfaces. 2016;8(21):13615-23.
- [101] Vafaei S, Tuck C, Ashcroft I, Wildman R. Surface microstructuring to modify wettability for 3D printing of nano-filled inks. Chem Eng Res Des. 2016;109:414-20.
- [102] Zhao H, Ye C, Fan Z, Shi Y. 3D printing of ZrO<sub>2</sub> ceramic using nano-zirconia suspension as a binder. Yarlagadda P. editor. Proceedings of the 2015 4th International Conference on Sensors, Measurement and Intelligent Materials; 2016. p. 654-7.
- [103] Manabe K, Nishizawa S, Shiratori S. Porous surface structure fabricated by breath figures that suppresses pseudomonas aeruginosa biofilm formation. ACS Appl Mater Interfaces. 2013:5(22):11900-5.
- [104] Zhang M, Zhang T, Cui T. Wettability conversion from superoleophobic to superhydrophilic on titania/singlewalled carbon nanotube composite coatings. Langmuir. 2011;27(15):9295-301.
- Fisher LE, Yang Y, Yuen M-F, Zhang W, Nobbs AH, Su B. [105] Bactericidal activity of biomimetic diamond nanocone surfaces. Biointerphases. 2016;11(1):011014.
- [106] Qin Y, Chen K, Gu W, Dong X, Lei R, Chang Y, et al. Small size fullerenol nanoparticles suppress lung metastasis of breast cancer cell by disrupting actin dynamics. J Nanobiotechnol. 2018;16:(1):54.
- [107] Peng F, Setyawati MI, Tee JK, Ding X, Wang J, Nga ME, et al. Nanoparticles promote in vivo breast cancer cell intravasation and extravasation by inducing endothelial leakiness. Nat Nanotechnol. 2019;14(3):279.
- [108] Yang G, Liu Y, Wang H, Wilson R, Hui Y, Yu L, et al. Bioinspired core-shell nanoparticles for hydrophobic drug delivery. Angew Chemie-International Ed. 2019;58(40):14357-64.
- [109] Garcia EB, Alms C, Hinman AW, Kelly C, Smith A, Vance M, et al. Single-cell analysis reveals that chronic silver nanoparticle exposure induces cell division defects in human epithelial cells. Int J Environ Res Public Health. 2019;16(11):2061.
- [110] Xie H, Wang P, Wu J. Effect of exposure of osteoblast-like cells to low-dose silver nanoparticles: Uptake, retention and osteogenic activity. Artif Cell Nanomed Biotechnol. 2019;47(1):260-7.
- [111] Tian X, Yang Z, Duan G, Wu A, Gu Z, Zhang L, et al. Graphene oxide nanosheets retard cellular migration via disruption of actin cytoskeleton. Small. 2017;13(3):1602133.
- [112] Shi X, Kong Y, Gao H. Coarse grained molecular dynamics and theoretical studies of carbon nanotubes entering cell membrane. Acta Mech Sin. 2008;24(2):161-9.
- [113] Gao Y, Mao D, Wu J, Wang X, Wang Z, Zhou G, et al. Carbon nanotubes translocation through a lipid membrane and transporting small hydrophobic and hydrophilic molecules. Appl Sci. 2019;9(20):4271.
- [114] Jameson CJ, Oroskar P, Song B, Yuan H, Murad S. Molecular dynamics studies of nanoparticle transport through model

- lipid membranes. In: Kök FN, Arslan Yildiz A, Inci F. Biomimetic lipid membranes: fundamentals, applications, and commercialization. Cham: Springer International Publishing; 2019. p. 109-65.
- [115] Lelimousin M, Sansom MSP. Membrane perturbation by carbon nanotube insertion: Pathways to internalization. Small. 2013;9(21):3639-46.
- [116] Luan B, Tien H, Zhao L, Zhou R. Potential toxicity of graphene to cell functions via disrupting protein-protein interactions. ACS Nano. 2015;9(1):663-9.
- [117] Hasan J, Webb HK, Vi Khanh T, Pogodin S, Baulin VA, Watson GS, et al. Selective bactericidal activity of nanopatterned superhydrophobic cicada Psaltoda claripennis wing surfaces. Appl Microbiol Biotechnol. 2013:97(20):9257-62.
- [118] Elbourne A, Chapman J, Gelmi A, Cozzolino D, Crawford RJ, Vi Khanh T. Bacterial-nanostructure interactions: The role of cell elasticity and adhesion forces. J Colloid Interface Sci. 2019;546:192-210.
- [119] Nowlin K, Boseman A, Covell A, LaJeunesse D. Adhesiondependent rupturing of Saccharomyces cerevisiae on biological antimicrobial nanostructured surfaces. J R Soc Interface. 2015;12(102):20140999.
- [120] Cheeseman S, Vi Khanh T, Walter V, Thalmann F, Marques CM, Hanssen E, et al. Interaction of giant unilamellar vesicles with the surface nanostructures on dragonfly wings. Langmuir. 2019;35(6):2422-30.
- [121] Bandara CD, Singh S, Afara IO, Wolff A, Tesfamichael T, Ostrikov K, et al. Bactericidal effects of natural nanotopography of dragonfly wing on Escherichia coli. ACS Appl Mater Interfaces. 2017;9(8):6746-60.
- [122] Xie Y, Qu X, Li J, Li D, Wei W, Hui D, et al. Ultrafast physical bacterial inactivation and photocatalytic self-cleaning of ZnO nanoarrays for rapid and sustainable bactericidal applications. Sci Total Environ. 2020;738:139714.
- [123] Michalska M, Gambacorta F, Divan R, Aranson IS, Sokolov A, Noirot P, et al. Tuning antimicrobial properties of biomimetic nanopatterned surfaces. Nanoscale. 2018;10(14):6639-50.

- [124] Ivanova EP, Hasan J, Webb HK, Vi Khanh T, Watson GS, Watson JA, et al. Natural bactericidal surfaces: Mechanical rupture of pseudomonas aeruginosa cells by cicada wings. Small. 2012;8(16):2489-94.
- [125] Linklater DP, Huu Khuong Duy N, Bhadra CM, Juodkazis S, Ivanova EP. Influence of nanoscale topology on bactericidal efficiency of black silicon surfaces. Nanotechnology. 2017:28(24):245301.
- [126] Pham VTH, Vi Khanh T, Orlowska A, Ghanaati S, Barbeck M, Booms P, et al. "Race for the Surface": Eukaryotic cells can win. ACS Appl Mater Interfaces. 2016;8(34):22025-31.
- Pham VTH, Vi Khanh T, Mainwaring DE, Guo Y, Baulin VA, Al Kobaisi M, et al. Nanotopography as a trigger for the microscale, autogenous and passive lysis of erythrocytes. J Mater Chem B. 2014;2(19):2819-26.
- [128] Alvarez R, Munoz-Pina S, Gonzalez MU, Izquierdo-Barba I, Fernandez-Martinez I, Rico V, et al. Antibacterial nanostructured Ti coatings by magnetron sputtering: From laboratory scales to industrial reactors. Nanomaterials. 2019;9(9):1217.
- [129] Ganjian M, Modaresifar K, Zhang H, Hagedoorn P-L, Fratila-Apachitei LE, Zadpoor AA. Reactive ion etching for fabrication of biofunctional titanium nanostructures. Sci Rep. 2019;9:18815.
- [130] Wandiyanto JV, Cheeseman S, Vi Khanh T, Al Kobaisi M, Bizet C, Juodkazis S, et al. Outsmarting superbugs: Bactericidal activity of nanostructured titanium surfaces against methicillin- and gentamicin-resistant Staphylococcus aureus ATCC 33592. J Mater Chem B. 2019;7(28):4424-31.
- [131] Zhang DE, Ni XM, Zheng HG, Li Y, Zhang XJ, Yang ZP. Synthesis of needle-like nickel nanoparticles in water-in-oil microemulsion. Mater Lett. 2005;59(16):2011-4.
- [132] Xiang Y, Li J, Liu X, Cui Z, Yang X, Yeung KWK, et al. Construction of poly(lactic-co-glycolic acid)/ZnO nanorods/ Ag nanoparticles hybrid coating on Ti implants for enhanced antibacterial activity and biocompatibility. Mater Sci Eng C Mater Biol Appl. 2017;79:629-37.