

Editorial

Special Issue: Advances in Engineered Nanostructured Antibacterial Surfaces and Coatings

Paul Cătălin Balaure 

“C.D. Nenițescu” Department of Organic Chemistry, Faculty of Chemical Engineering and Biotechnologies, University Politehnica of Bucharest, 011061 Bucharest, Romania; pbalaure@gmail.com or paul.balaure@upb.ro

Pathogenic biofilm formation is a major issue of concern in various sectors such as healthcare and medicine, food safety and the food industry, wastewater treatment and drinking water distribution systems, and marine biofouling [1–3]. Biofilms are sessile bacterial colonies attached to an abiotic or biotic surface and sheltered by a matrix of self-produced extracellular polymeric substances (EPS), namely proteins (e.g., fibrin), polysaccharides (e.g., alginate), and eDNA [4]. Despite being genetically identical, in the biofilm style of life, bacteria exhibit an altered phenotype with respect to metabolic activity, rate of growth, and gene expression as compared to their planktonic counterparts [5]. Even cells located in different regions of the EPS matrix of the same biofilm experience different local conditions regarding nutrients and oxygen shortages as well as increased levels of waste products, secondary metabolites, and secreted factors [6,7]. As a normal consequence, gene expression in these cells as well as in their free-floating counterparts differ significantly. In biofilm aggregates, bacterial cells communicate with each other, somehow similar to multicellular organisms, which enables them to behave like a group in a coordinated and intricately regulated manner through a mechanism known as quorum sensing (QS) [8]. This intercellular communication is mediated by signaling molecules that control the biofilm formation and development, namely N-acyl-homoserine lactones (AHL) or auto-inducing peptides (AIP) in Gram-negative and Gram-positive bacterial pathogens, respectively [9–13]. Moreover, the developed communication abilities provide bacteria with a lot of benefits in host colonization and self-defense against antimicrobials and host immune system resulting in increased recalcitrance and resilience towards the most commonly used agents in the anti-infective therapy, which explains the chronic and recurrent character of biofilm infections. Combined with the selection of multidrug resistant strains as a result of excessive use and misuse of antibiotics, an acquired pathogenic biofilm infection can become a life-threatening condition, especially in the case of nosocomial infections associated with the use of invasive, interventional, indwelling, and implanted medical devices [14]. Covering of surfaces with nanostructured biofilm resistant coatings appeared as a valuable effective way to address this serious public health problem.

Two main strategies emerged in designing an anti-biofilm nanocoating, the so-called passive and, active strategies.

The passive strategy aims to impede the settlement of bacteria on a surface (“fouling resistance” approach), or to remove the already attached bacteria (“fouling release” approach) [13,15]. Surface modulation of bacterial adhesion underlying the passive strategy has to take into account a delicate balance between several surface parameters that influence adhesion-like surface topography, namely surface roughness and micropores, physical properties of the substrate—i.e., the mechanical stiffness and wettability based on the molecular hydrophilic/lipophilic balance—as well as the chemical properties of the surface such as surface energy, surface charge, and the presence of bioactive molecules on the substrate [16]. Fouling resistance coatings are obtained by the construction of a



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hydrophilic surface numerous hydrophilic materials, such as poly(ethylene glycol), zwitterionic, glycomimetic, and peptidomimetic polymers being employed to this purpose. The mechanism behind the antifouling effect is based on thermodynamic considerations: (a) the steric repulsion effect originating in the unfavorable entropy loss occurring when a foulant compresses the extended polymer brushes thereby restraining the free motility of the polymer chains, and (b) non-specific foulant adsorption is thermodynamically disfavored since it would involve disruption of the strongly bound hydration shell present at the hydrophilic surface [13,15]. On the other hand, the fouling release approach was inspired by the low adhesion and self-cleaning properties of the Lotus leaf resulting from a peculiar surface topography with dual hierarchical roughness at the micro- and nano-length scales consisting of micro-bumps of epidermal cells (papillae) coated with a dense covering of agglomerated nanoscale epicuticular hydrophobic wax tubules [17]. To construct such low adhesion coatings, low surface energy polymer materials such as silicones and fluoropolymer derivatives were mostly used [18,19], but metallic and metal oxides anti-biofouling surfaces with nanostructured hierarchical topography have been also developed [20]. Combination of the two passive strategies in a unique platform in order to mitigate the inherent drawbacks associated with each of them and to achieve a synergic antifouling effect was reported as well [21]. Several methods to fabricate superhydrophobic coatings with hierarchical roughness have been developed. From the top-down fabrication techniques, we mention: (1) templating, which is a replication technique allowing the creation of a surface topography that is a complementary (negative) replica of an appropriate specially prepared template [22,23]; (2) lithography, including photolithography, electron beam lithography, and ion beam lithography, which all share the principle of transferring an image from a mask to a receiving substrate [24,25]; (3) chemical and plasma etching [26,27]; (4) anodic oxidation [28]; and (5) laser ablation [29]. In bottom-up methods, the nanopatterning of the surface is achieved by the controlled sequential deposition of material onto a substrate. The deposition techniques can be based on (1) purely physical processes such as evaporative methods, physical vapor deposition with its laser-assisted variants pulsed laser deposition (PLD) and matrix-assisted pulsed laser evaporation (MAPLE), and electrospinning [30,31]; (2) physicochemical processes such as molecular self-assembly [32,33]; and (3) chemical processes involving a chemical reaction between appropriate precursors. With regard to the latter category, the precursors may initially be either in solution this being the case of sol-gel [34], hydro- or solvothermal [35], and electrochemical [36–38] deposition processes, or in the vapor phase like in the chemical vapor deposition (CVD) method [39].

As the opposite of the antibiofouling passive strategies [15], active strategies aim either to kill bacteria by destroying vital cellular structures such as the plasma membrane or by interfering with crucial metabolic processes or to mitigate the increase in recalcitrance and virulence associated with biofilm development by disrupting the molecular mechanisms and signaling pathways that control the different stages of the biofilm life cycle. There are two types of active antibiofilm coatings, namely contact killing or non-leaching and drug eluting. The therapeutic agent can be either covalently tethered on the coating's surface (contact killing) or entrapped mechanically or via intermolecular bonding within the coating's bulk acting like a drug reservoir which is gradually depleted (drug eluting) [40–43]. Structurally, there are several types of nano-coatings such as self-assembled monolayers (SAMs), multi-layered layer by layer (LbL) deposited coatings, polymer brushes or ionotropically cross-linked hydrogels [44]. Nanostructured active antibiofilm coatings can target all main mechanisms responsible for the acquired increased recalcitrance of biofilm bacteria towards antibiotics, namely alteration of cell membrane permeability resulting in decreased drug uptake and increased drug export, enzymatic destruction of the antibiotic molecule, and the modification/absence of the antibiotic targets [45,46]. There is a wide variety of antimicrobial agents: antibiotics of various classes and mechanisms of action, antimicrobial peptides, antimicrobial enzymes, quorum sensing inhibitors, efflux pumps inhibitors, nucleotide second messenger signaling modulating molecules, persister cell formation inhibitors, quorum quenching agents, biofilm dispersal inducers, bacterial genetic

biodiversification inhibitors, polycationic biocides, N-halamine compounds, chlorhexidine, usnic acid, silver and silver ions, and a series of natural products such as resveratrol and essential oils [3,12,46].

Advanced smart engineered nanocoatings fulfill a series of key requirements needed especially for biomedical applications [47].

First, we will discuss the striking advantageous features of multi-task drug releasing nanocoatings.

- A. Modulation of release kinetics in order to keep the concentration of the antimicrobial agent within the therapeutic window as long as necessary can be achieved using polyelectrolyte multi-layered nanocoatings (PEMs) formed by LbL deposition of oppositely charged polymers [48].
- B. Smart nanocoatings which are able to undergo structural changes in sharp response to particular endogenous (e.g., small changes in microenvironmental temperature, pH, enzyme activity, or redox potential) or exogenous externally applied (electrical, ultrasonic, photothermal, magnetic, and mechanical) stimuli, thereby triggering the release of their therapeutic payload [49]. The most advantageous variant of this approach is represented by the bacteria-responsive coatings, which release their drug payload only when surrounded or in contact with bacteria [50], thereby mitigating unwanted side effects, resistance development, and futile drug use.
- C. Multi-release coatings which can simultaneously deliver different antimicrobials with different action mechanisms. The aim of this approach is to achieve synergic effects and to reduce induction of bacterial resistance since several mutations should occur simultaneously in the same bacterial cell.
- D. Multi-property coatings fulfil a series of requirements which are of primordial importance in clinical applications such as biocompatibility, lack of toxicity and immunogenicity, mechanical strength, resistance to corrosion and wear, anticoagulation, enhanced bone-integration, and improved overall tissue-integration [47].

Second, the latest trends in the field of antibiofilm coatings refer to the so-called multi-approach coatings, which join in a unique nanoplatform both contact killing and drug eluting strategies. In operating these unique integrating platforms, the two strategies can be applied either simultaneously or sequentially, that is, one at a time. In the latter case, depending on the initial status of the nanocoating, two strategies emerged, namely the “kill and repel” and the “resist and kill” surfaces [51]. One of the “kill and repel” approaches is based on a pH-induced chemical switching strategy from an initial antibacterial cationic form of the surface to a fouling resistance zwitterionic form [52]. On the other hand, the “resist and kill” approach is illustrated here by the following example [53]. A multi-layered coating was constructed by LbL self-assembly of alternate layers of oppositely charged polyanionic heparin and polycationic chitosan. At the top of this coating another multi-layered coating of cross-linked polyvinylpyrrolidone/poly(acrylic acid) was deposited by hydrogen bond interactions. The highly hydrated and thus fouling resistant top coating undergoes controlled degradation in a predetermined time frame (24 h), living behind the outermost polycationic and thus contact killing chitosan layer of the inner multi-layered coating [53]. By making the switching process from the initial bactericidal status to the non-fouling status reversible, self-healing or self-regenerating “kill and repel” surfaces, which can go through several bacteria killing/surface regeneration cycles without significant reduction in the initial bactericidal activity, could be obtained [40,44,54,55].

The aim of the present Special Issue is to highlight the latest achievements in the field of engineered nanostructured coatings, regardless of structure and morphology, chemical composition, action mechanism or fabrication method, but with a special focus on the applications in the biomedical and food industry fields.

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