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Editorial

Special Issue: Biointerface Coatings for Biomaterials and Biomedical Applications

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The success of recent material science and applications in biotechnologies should be credited to developments of malleable surface properties, as well as the adaptation of conjugation reactions to the material surface [1–4]. An article additionally addressing the progress and challenges of biointerface modifications using integrated nanomaterial bioimplants for orthopedic applications was also thoroughly reviewed in the current Special Issue by Ahirwar et al [5]. A biointerface is the region of contact between a biomolecule, cell, biological tissue, or living organism considered living with another biomaterial or inorganic/organic material. The surface property of a material is highly important, even compared to the bulk property [6]. For example, a hydrogel is soft (e.g., 1–100 kPa) but could have a stiffer thin layer on the top surface (>100 kPa) that modulates cell adhesion at the biointerface. On the other hand, rigid materials can also be modified with a softer surface coating to enhance biocompatibility. Therefore, surface modification or coatings have immense potential and wide application in biomedical devices and implants [7]. The crucial biointerface properties of using biomaterials include the ability to control the presentation of biomolecules with precisely defined chemical topology, the ability to control and suppress undesirable background noise from nonspecific biomolecules (e.g., protein) adsorptions, smart response with respect to environmental stimuli, multiple functions that are simultaneously activated, and surface gradients with gradual and cascade guidance from physical and/or chemical cues. These properties may subsequently lead to successful biodevice/material performance and efficacy. In the article "*Proteomic Analysis of Biomaterial Surfaces after Contacting with Body Fluids by MALDI-TOF Mass Spectroscopy*", Hirohara et al. developed an analysis method to evaluate the composition of protein absorbed on solid surfaces while preventing the effects of pipetting artifacts. The method includes denaturation, reduction, alkylation, digestion, and spotting of the matrix followed by matrix-assisted laser desorption/ionization-time of flight mass (MALDI-TOF mass) spectroscopy. The authors also developed an algorithm to evaluate the adsorbed proteins by collating the experimental and theoretical peak positions of fragmented peptides. Their results showed the mechanism of how the cell and tissues are affected after biomaterial contact [8].

Furthermore, biomolecular engineering technologies have enabled the successful use of these polymer materials. The use of physical approaches or chemical means to install biological functions onto polymer materials is of interest within this area of research. From a physical point of view, the ability to control biomolecules at the solid/liquid interface requires adequate knowledge and understanding of surface interactions, transport phenomena of interacting molecules, interactions with external stimuli, and surface functional groups. From a chemical point of view, conjugation reactions seek a fast reaction time, mild reaction conditions, and, more importantly, specificity to achieve successful conjugation in the vast array of functionalities present in biological microenvironments [9].

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Recently demonstrated and promising concepts have focused on the creation of multiple surface functionalities on material surfaces [10–15]. The approaches consider the physical and chemical surface properties while delivering cascading and/or simultaneous activities to respond to sophisticated bioenvironments. Moreover, the need to precisely incorporate biomolecules at specific locations on a micro/nanoscale, i.e., in confined micro/nanodomains and to induce topographically derived responses toward biological environments, has also become essential. These concepts have fueled modern schemes for the design of prospective polymer materials for biological applications as well as more advanced developments in biointerface science. In the article "*Facile Route of Fabricating Long-Term Microbicidal Silver Nanoparticle Clusters against Shiga Toxin-Producing Escherichia coli O157:H7 and Candida auris*", Gangadoo et al. reported a facile coating technique using silver nanoparticles (Ag NPs) to fabricate a surface with long-term microbiocidal activity. The Ag NP coating was fabricated on copper surfaces via an ion-exchange and reduction reaction followed by a silanization step, resulting in high-aspect-ratio Ag NP clusters. Their results demonstrated the durability and high efficiency of microbiocidal activity against both Shiga toxin-producing *E. coli* and *C. auris* cells [16]. In the article "*One-Step Preparation of Nickel Nanoparticle-Based Magnetic Poly (Vinyl Alcohol) Gels*", Li et al. established a one-step synthetic method of magnetic poly (vinyl alcohol) (mPVA) gels. Using this method, multiresponsive gels were obtained by incorporating Ni nanoparticles (NPs) into a stimuli-responsive polymer. Ni-NP PVA gels are anticipated to be applied for controlled drug delivery systems, especially anticancer applications. This novel and facile method can be utilized for magnetic gels for biotechnology [17]. In the article "*Mangrove Inspired Anti-Corrosion Coatings*", Cui et al. discovered that well-controlled transport of corrosive substances is the critical key to anti-corrosion performance. The authors demonstrated a bipolar hydrophobic coating that can effectively block and control the transport of both Na + and Cl-, exhibiting significantly improved anti-corrosion properties and a more than three orders of magnitude decrease in corrosion current density. These bioinspired coatings may lead to outstanding and long-term anti-corrosion performance [18].

Surface topography, including nanostructure and roughness, is one of the critical parameters of materials at biointerfaces. Advanced technologies, including electron beam lithography, laser ablation, and electrochemical etching, have been developed to generate surface nanotopographies [19,20]. In the article "*Vapor-Stripping and Encapsulating to Construct Particles with Time-Controlled Asymmetry and Anisotropy*", Wu et al. demonstrated the fabrication method of particles with asymmetric and anisotropic structures by utilizing a time-controlled vapor stripping and encapsulating process. The results showed that the innovative process, chemical vapor sublimation and deposition (CVSD), enabled sensitive soybean agglutinin (SBA) protein tubes to be encapsulated in poly-p-xylylene particles. The SBA protein tubes retained their original morphology and could be used to construct particles with asymmetric and anisotropic structures. In addition, the size of the particles could be predicted and controlled. The CVSD process is a promising strategy for fabricating particles [21]. Nevertheless, these methods are material-dependent. For example, silicon has been commonly used in lithography and etching methods. Nanostructures have been used to control cell adhesion and differentiation. The outcomes depend on the size, geometry, arrangement, density, and, not surprisingly, cell type. In general, nanostructures are biocompatible, cell adhesive, low bacterial adhesive, low inflammatory, and biofunctional. For example, nanostructures on a bone implant should induce a mild inflammatory effect and good bone cell affinity and bone integration. In the article "*Icariin/Aspirin Composite Coating on TiO_2 Nanotubes Surface Induce Immunomodulatory Effect of Macrophage and Improve Osteoblast Activity*", Ma et al. used an aspirin (ASP)/poly (lactic–co–glycolic acid) (PLGA) coating on icariin (ICA)-loaded TiO_2 nanotubes (NT-ICA-ASP/PLGA). Compared to those cultured on the Ti surface, macrophages on the NT-ICA-ASP/PLGA substrate displayed decreased M1 proinflammatory and enhanced M2 pro-regenerative gene and protein expression, which implied an activated immunomodulatory effect. Moreover, when cultured with conditioned medium from macrophages, osteoblasts on the NT-ICA-ASP/PLGA

substrate showed improved cell proliferation, adhesion and osteogenic gene and protein expression compared with those on the Ti surface. These results suggested that the NT-ICA-ASP/PLGA substrate is a promising candidate for functionalized coating material in Ti implant surface modification [22]. In the article "*The Bioactive Polypyrrole/Polydopamine Nanowire Coating with Enhanced Osteogenic Differentiation Ability with Electrical Stimulation*", He et al. used a two-step method to construct a functional conductive coating of polypyrrole/polydopamine (PPy/PDA) nanocomposites for bone regeneration. The PPy/PDA NW coating exhibited better biocompatibility and bioactivity than pure PPy NWs and PDA and was beneficial for the adhesion, proliferation, and osteogenic differentiation of MC3T3-E1 cells cultured on the surface. In addition, PPy/PDA NWs significantly promoted the osteogenesis of MC3T3-E1 cells in combination with micro galvanostatic electrical stimulation (ES) [23].

Surface chemistry, including material chemistry, biomolecule grafting, and chemical coatings, is another critical parameter of materials at biointerfaces [24–26]. Surface chemistry represents functional groups, electrostatic properties, and wettability on the material surface. These properties are crucial in interactions with proteins, cells, and tissues. For example, the carboxy group can stimulate osteogenic differentiation of mesenchymal stem cells (MSCs). Uniform coatings such as spin coating provide a thin layer of polymer (~submicrons) on materials. The adhesive force between the thin layer and substrate is essential for the long-term application of the materials. Nonuniform coatings can be achieved using dewetting methods, which in turn generate nano or micrometer-scale features. In the article "*Corrosion Behavior and Biological Activity of Microarc Oxidation Coatings with Berberine on a Pure Magnesium Surface*", Mu et al. reported a coating named ultrasonic microarc oxidation/polylactic acid and glycolic acid copolymer/berberine (UMAO/PLGA/BR) on a pure magnesium substrate for bone materials. Different amounts of berberine (BR) can seal the corrosion channel to different extents. These coatings have a self-corrosion current density (Icorr) at least one order of magnitude lower than that of the UMAO coatings. When the BR content was 3.0 g/L, the self-corrosion current density of the UMAO/PLGA/BR coatings was the lowest (3.14×10^{-8} A/cm^2), and the corrosion resistance was improved. UMAO/PLGA/BR coatings have excellent biological activity, which can effectively solve the clinical problem of rapid degradation of pure magnesium and easy infection [27]. In the report "*Chemical and Biological Roles of Zinc in a Porous Titanium Dioxide Layer Formed by Micro-Arc Oxidation*" by Shimabukuro et al., zinc incorporated by microarc oxidation (MAO) in porous titanium dioxide was investigated under physiological saline conditions. The time transient state from zinc to zinc oxide led to early stage release in 7 days and antibacterial ability after 28 days of incubation. Additionally, there was no interruption of osteogenic cell proliferation and calcification in zinc specimens. In conclusion, time-transient zinc not only gives antibacterial properties but also shows great compatibility with osteogenic cells and has great potential in chemical and biological fields [28].

Surface stiffness in the material top surface (~submicrons) is crucial in controlling cell adhesion, the cytoskeleton, and cell differentiation [29]. In vivo, the stiffness of the extracellular matrix (ECM) ranges from a few kPa (e.g., brain) to GPa (e.g., bone). Thus, it makes sense that surfaces with different stiffnesses could mimic the native ECM and guide cells to become a specific cell type. The top surface of materials is easily oxidized, altering the stiffness of the surface. Because the top surface has a more dominant effect on cell behavior than the bulk material, technology with higher resolution is needed to analyze the mechanical properties of the top layer. In the article "*Mechanical Properties of Strontium–Hardystonite–Gahnite Coating Formed by Atmospheric Plasma Spray*", Pham et al. measured the mechanical properties and tested the cell viability of a bioceramic coating, strontium–hardystonite–gahnite (Sr–HT–G, Sr–$Ca_2ZnSi_2O_7$–$ZnAl_2O_4$), to evaluate the potential use of this novel bioceramic for bone regeneration applications contrasted to the properties of the well-known commercial standard coating of hydroxyapatite (HAp: $Ca_{10}(PO_4)_6(OH)_2$). The Sr–HT–G coating exhibited a more uniform distribution of hardness and elastic moduli across its cross-section compared to HAp. The Sr–HT–G coating also

revealed higher microhardness, nanohardness and elastic moduli than those shown for the HAp coating. The nanoscratch tests for the Sr–HT–G coating presented a low volume of material removal without high plastic deformation. Furthermore, the Sr–HT–G coating had a lower wear volume than the HAp. The Sr–HT–G coating had a slightly higher cell attachment density and spreading area of bone marrow mesenchymal stem cells (BMSCs) than the HAp coating [30].

Finally, biointerfaces often contain a combination of surface topography, chemistry, and stiffness. These three properties are sometimes difficult to distinguish. For example, the generation of surface nanotopography could change the surface wettability, and polymer coating could alter the surface nanotopography. Recently, a new family of two-dimensional (2D) materials called colloidal self-assembled patterns (cSAPs), composed of different particles, has been developed [31,32]. The formed particle patterns can be hexagonal, close-packed, or randomly distributed. The topography of cSAPs can be tuned by controlling particle-particle interactions. It is easy to decorate biosignals on cSAPs using pre- or postmodification of particles. cSAPs can be used as substrates, coatings, and free-standing membranes, depending on the application. cSAPs can be the next-generation material at biointerfaces. As more stringent specifications are required for designing the surface properties of prospective materials and the development of new devices is pursued with complicated geometries and minimized sizes, the surface properties of such materials/devices now also require a more defined and flexible presentation of the chemical functionalities (e.g., multifunctional or gradient distribution) and the precise confinement of these chemical conducts in relevant locations of interest [33]. The emerging applications of the existing technologies and/or new technologies from two dimensions into more sophisticated three-dimensional regions [34] are challenging the field of biointerfaces science, and further research developments are expected on this path.

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