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Mussel-inspired polydopamine for bio-surface functionalization

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Abstract

Surface functionalization via molecular design has been a key approach to incorporate new functionalities into existing biomaterials for biomedical application. Mussel-inspired polydopamine (PDA) has aroused great interest as a new route to the functionalization of biomaterials, due to its simplicity and material independency in deposition, favorable interactions with cells, and strong reactivity for secondary functionalization. Herein, this review attempts to highlight the recent findings and progress of PDA in bio-surface functionalization for biomedical applications. The efforts made to elucidate the polymerization mechanism, PDA structure, and the preparation parameters have been discussed. Interactions between PDA coatings and the various cell types involved in different biomedical applications including general cell adhesion, bone regeneration, blood compatibility, and antimicrobial activity have also been highlighted. A brief discussion of post-functionalization of PDA and nanostructured PDA is also provided.
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Keywords: Polydopamine; Functionalization; Biomedical application; Polymerization

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1. Introduction

Surface modification and functionalization play a central role in controlling surface properties and conferring new functionalities to materials, which is especially important in critical fields such as biomaterials, tissue engineering, and medical diagnostics [1–3]. In the past few decades, several key technologies have been developed to modify material surfaces, such as chemical conjugation [4], self-assembly monolayers (SAMs) [5], layer-by-layer (LBL) film deposition [6], and plasma treatment [7]. In general, however, most these surface modification techniques are time consuming and complicated processes, and, more critically, their application and quality rely largely on specific surface properties. For instance, SAMs of thiolates can only form stable layers on noble metals [5,8]; while alkylsilane SAMs can only be applied to silicon dioxide and silicon surfaces [9]. Therefore, a simple and universal surface modification approach with wide substrate applicability and easy processing is highly desirable.

In nature, mussels adhere strongly to wood or stones in wet conditions of high shear stresses from water flow. How can the mussel be an underwater specialist? The secret lies in the different mussel foot proteins secreted during adhesive formation in the adhesive plaque of mussel byssus. It was found that all these foot proteins contain 3,4-dihydroxy-L-phenylalanine (DOPA) and lysine amino acids [10,11], leading to the hypothesis that the co-existence of catechol (DOPA) and amine (lysine) groups may be crucial for achieving strong adhesion [12]. Polydopamine (PDA), containing both catechol and amine groups, was discovered to be a one-step facile surface coating method in 2007 [12]. The PDA coating has been demonstrated to functionalize a wide array of material surfaces, including superhydrophobic surfaces. Hence, PDA has opened a new route for surface modification and has garnered great interest, especially in materials science, biology and biomedical fields.

The aim of this review article is to outline the recent findings of PDA buildup mechanism and surface properties, and the development of PDA films as a smart coating material for biomedical applications. In the first part of this review, we will summarize what has been found regarding the buildup mechanisms and proposed structures of the PDA film as well as parameters that influence the film formation. In the second part, research involving the interactions of PDA coatings with mammalian cells and bacteria as well as the underlying mechanism of cell responses will be discussed. Finally, we will further present some representative applications of PDA as multifunctional coating in the biomedical field.

2. Coating mechanism, structure, and preparation

2.1. Coating mechanism and structure

The PDA is spontaneously formed by pH-induced, oxidative polymerization of dopamine-hydrochloride in alkaline solutions (pH $>$ 7.5). To achieve PDA coatings, simple immersion of substrates in a dilute aqueous solution of dopamine (typically 2 mg/mL of dopamine in 10 mM TRIS buffer) results in spontaneous deposition of a thin PDA film [12]. Despite the ease of the PDA coating preparation, the molecular mechanism of the PDA buildup is still not fully understood, which is mainly due to the marked chemical heterogeneity and adverse physical properties of this material. Further, PDA structure can depend on the reaction conditions, e.g. buffer solution [13–15], and thus the discussion here will focus on PDA obtained by most commonly used protocol, i.e. air oxidation in TRIS or phosphate buffer.

In solution, it is well known that PDA buildup shares the first steps with melanin biosynthesis: the oxidation of dopamine to dopamine-quinone, its intramolecular cyclization, oxidation to dopaminechrome, formation of 5,6-dihydroxyindole (DHI), and further oxidation to 5,6-indolequinone (IDQ) (Scheme 1) [16]. It is notable that the mixture of the dopamine, quinone and indole units may co-exist in solution after the first steps since the oxidation and cyclization may not be complete. Subsequently, the mixture of these units undergoes various pathways to form the PDA structure, which presents a puzzle: is PDA a covalent polymer or a supramolecular aggregate?

In 2007, Messersmith and co-workers examined the mass spectra of PDA-coated glass using time-of-flight secondary ion mass spectrometry and found a peak at *m/z* 445 which was assigned to the DHI trimer [12]. Based on this observation, they suggested a polymerized structure (Scheme 1 I) wherein the DHI molecules undergo branching reactions at positions 2, 3, 4, and 7, leading to multiple isomers of dimers and later on higher oligomers, which eventually form the covalent PDA structure. However, the peak at *m/z* 445 was not present in the time-of-flight secondary ion mass spectrometry spectra of the PDA-coated substrates fabricated with the same protocol in other studies [17–19]. Later, Liebscher and co-workers reported investigations of the PDA structure using various spectroscopic methods, e.g. solid-state nuclear magnetic resonance, electrospray ionization high-resolution mass spectrometry, X-ray photoelectron spectroscopy [20]. Here the authors demonstrated that PDA was a covalent polymer while the buildup units consisted of mixtures of various indole units with different degrees of (un)saturation and open-chain dopamine units, rather than a single DHI unit (Scheme 1 II).

In contrast to the covalent polymer models, Bielawski and co-workers analyzed the PDA structure using a variety of solid state spectroscopic and crystallographic techniques [21]. Their data revealed the presence of hydrogen bounds to the aryl core of the PDA and stacked structures formed by monomers. Therefore, they proposed that PDA is not a covalent polymer but instead a supramolecular aggregate of monomers (primarily DHI and its dione derivative) that were held together via a combination of charge transfer, π-stacking, and hydrogen bonding interactions (Scheme 1 III). In the other study [22], Lee and co-workers monitored the PDA formation by high-performance liquid chromatography mass spectrometry and identified that a physical, self-assembled trimer of (dopamine)$_2$/DHI (Scheme 1 IV) exists in PDA.

The proposed model was further advanced by d'Ischia and co-workers in a recent report [15]. Here the authors demonstrated that PDA consists of three main building blocks, uncyclized catecholamine/quinones, cyclized DHI unites, and

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