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## Polydopamine Surface Chemistry: A Decade of Discovery

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### Abstract

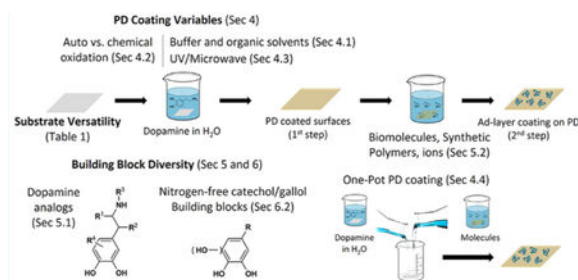
Polydopamine is one of the simplest and most versatile approaches to functionalizing material surfaces, having been inspired by the adhesive nature of catechols and amines in mussel adhesive proteins. Since its first report in 2007, a decade of studies on polydopamine molecular structure, deposition conditions, and physicochemical properties have ensued. During this time, potential uses of polydopamine coatings have expanded in many unforeseen directions, seemingly only limited by the creativity of researchers seeking simple solutions to manipulating surface chemistry. In this review, we describe the current state of the art in polydopamine coating methods, describe efforts underway to uncover and tailor the complex structure and chemical properties of polydopamine, and identify emerging trends and needs in polydopamine research, including the use of dopamine analogs, nitrogen-free polyphenolic precursors, and improvement of coating mechanical properties.

### Graphical abstract:

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## 1. INTRODUCTION

Polydopamine (PD) is a uniquely adaptable and simple surface functionalization method, being the first single-step, material-independent surface chemistry when it was first reported in 2007.<sup>1</sup> Since its introduction, PD has emerged as one of the most powerful tools available for modification of surfaces, achieving this status as a result of versatility, simplicity, and broad potential use in the biomedical, energy, consumer, industrial, military, and other sectors. In this review, we will summarize the evolution of the PD method since it was first reported ten years ago. We begin by describing the origin and basic features of PD, which arose as an outcome of mussel adhesive protein research. Next, we compare PD coatings to other commonly employed surface modification methods, followed by an account of how approaches to PD deposition have evolved from the original “recipe” to the current state-of-the-art PD preparation methods. We then provide a synopsis of the substrates and materials found to be amenable to modification with PD, and introduce selected emerging applications for these coatings. Finally, we conclude with a forward-looking statement on the opportunities and challenges in further development and implementation of PD and PD-like coatings.

## 2. INSPIRATION AND GENERAL FEATURES OF PD

The invention of PD originated from previous studies on one of Nature’s most celebrated families of wet adhesive biomolecules- the mussel adhesive proteins. These proteins, especially the *mytilus* foot proteins-3 and -5 (Mfp-3 and -5) located in the distal portion of the mussel byssus where the byssal foot engages the substrate surface,<sup>2-4</sup> have two key features that inspired PD: (1) high catechol (3,4-dihydroxybenzene) content due to the presence of 3,4-dihydroxy-L-phenylalanine (DOPA); and (2) high primary and secondary amine content due to lysine (Lys) and histidine residues. The high concentration and intimate association of DOPA and Lys/ His was noted early on by Waite and co-workers as a remarkable feature of Mfp-5,<sup>3</sup> leading to speculation that the combination of catechol and amine is a special one as it relates to interfacial adhesion. Early exploitation of catecholamines as building blocks for bioinspired materials include the synthesis of DOPA-Lys poly(amino acids),<sup>5</sup> the tethering of catechol moieties to amine-rich polymers such as poly(ethylenimine)<sup>6,7</sup> and chitosan<sup>8,9</sup> and the use of short DOPA-Lys peptides as anchors for

immobilization of antifouling polymers at solid–liquid interfaces.<sup>10</sup> Although low molecular weight catecholamines such as dopamine were not considered in the context of adhesion prior to 2007, the coexistence of catechol and amine functional groups that is such a distinctive feature of mussel adhesive proteins is now understood to be a powerful combination (only recently was the physicochemical basis for the interfacial adhesive synergy between catechol and amine investigated in detail<sup>11</sup>).

The widespread adoption of PD originates from its simplicity, low cost and adaptability in a variety of science and applied engineering contexts. While variations of the coating method exist as discussed below, in its simplest manifestation coating an object with PD involves nothing more than simply immersing it in an aqueous alkaline solution of dopamine for an adjustable period of time (Figure 1). Spontaneous deposition of a conformal PD coating occurs during incubation, and this primary coating can be used without further modification or used as a “primer” onto which a subsequent secondary coating is applied. The composition and properties of the secondary coating is highly tailorable, therefore giving rise to the tremendous versatility and broad range of applications enjoyed by PD coatings. Dopamine-HCl is a commercially available and relatively inexpensive reagent (3.2 USD per gram when purchasing from Sigma-Aldrich). For little cost, one can make a one-liter solution of dopamine (1 mg/mL) that can be sprayed<sup>12</sup> or used as an aqueous bath for dip-coating large surface areas.

The formation of PD coatings occurs by oxidative polymerization of dopamine, the details of which remain an active area of investigation. In fact, many features of PD formation and structure remain unknown. For this reason, and because other research and review papers with detailed mechanisms have been recently published,<sup>13–19</sup> we will provide only a brief overview of existing theories of PD formation and structure here (Figure 2). There is little doubt that the initial driving force for PD formation is the oxidation of dopamine by dissolved oxygen at alkaline pH of the solution, as elimination of oxygen from the solution slows or eradicates PD formation. The oxidation product, dopamine-quinone, undergoes a nucleophilic intramolecular cyclization reaction leading eventually to the formation of 5,6-dihydroxyindole. In most existing theories of PD formation, these two compounds, dopamine-quinone and 5,6-dihydroxyindole (DHI), are key building blocks for PD, albeit through various proposed pathways. On one hand, it has been postulated that PD is composed entirely of noncovalent assemblies of dopamine, dopamine-quinone and DHI, whereas other hypotheses hold that these molecules polymerize to form a heteropolymer composed of catecholamine, quinone and indole repeat units. Alternatively, it has been suggested that PD is a eumelanin-like material composed of oligomeric building blocks generated spontaneously by further oxidation of DHI and coupling through 2–2', 4–7', 2–4', and/or 2–7' linkages. It should be mentioned that there is currently no consensus on the PD formation mechanism, and the proposed noncovalent and covalent pathways elaborated above should not be viewed as mutually exclusive, as it is possible and perhaps even probable that both covalent “polymerization” and “self-assembly” pathways contribute to PD formation.<sup>20</sup> In addition, M. d'Ischia and co-worker found pyrrolecarboxylic acid (PCA), an oxidative degraded form of PD, with uncyclized dopamine/dopamine-quinone and cyclized DHI units when forming PD (Figure 3a).<sup>17</sup> Ding et al. reported that (DHI) /PCA trimer complex can also be one of building blocks of PD<sup>18</sup>. This trimer is

associated with others by noncovalent interactions to form the PD (Figure 3b). Additional uncertain aspects of PD formation that should be addressed by future studies, relate to how the aforementioned events occurring in solution lead to spontaneous deposition of a conformal coating on solid surfaces, and how the composition of the coating is the same as the product that can be isolated from the surrounding solution.

### 3. PD IN COMPARISON TO OTHER COATING METHODS

The unique ability of PD to be deposited as a conformal thin film onto virtually all types, shapes and sizes of organic and inorganic surfaces through a simple dip coating process distinguishes it from other approaches to surface modification. Before the discovery of PD coatings, the three dominant methods in surface modification chemistry were self-assembled monolayer (SAM),<sup>21–23</sup> layer-by-layer (LbL) assembly,<sup>24,25</sup> and plasma treatment.<sup>26–28</sup> In SAMs, end-functionalized alkanethiol molecules form ordered monolayers on noble metal surfaces through highly specific metal–thiolate bonds<sup>22</sup> and therefore require matching surface–adsorbate chemistries. On the other hand, gas-phase plasma surface chemistry modifications are only transient because the modified surface properties change with time.

It is worthwhile to briefly compare PD and LbL coating methods (Figure 1), as both methods are arguably the most versatile and rely on adsorption of coating components at solid–liquid interfaces. Despite some similarities, there remain some important differences between PD and LbL coatings in terms of mode-of-action, properties, and substrate versatility. First, PD in its simplest form is a synthesis-free method in which the coating can be built in one step without the need to procure or synthesize sophisticated polymers or other coating components (ad-layer components notwithstanding). In contrast, polymers used in LbL methods are often synthesized specifically with a view toward LbL use or to provide a new function, and the deposition processes often involve many coating cycles. Second, formation of PD coatings involves in situ polymerization starting from its “monomer” dopamine, which is covalently/noncovalently polymerized at later stages (see above). Therefore, PD coatings are ideally suited for coating of 3D porous materials by infusion and in situ polymerization due to the low molecular weight of the dopamine building block. In contrast, polymers used in LbL assembly are typically high molecular weight and therefore will diffuse rather slowly into 3D substrates with small pores. Third, thickness of layers formed by LbL assembly can be easily controlled from a few nanometers to several micrometers by varying the number of deposition cycles, which in some cases can be hundreds of cycles. However, PD deposition is a kinetic process with a saturation limit of typically less than 50 nm due to dopamine depletion from the coating solution (it is possible to deposit additional PD layers onto a pre-existing PD layer). Fourth, the PD layer exhibits intrinsic chemical reactivity originating from the presence of catecholquinone moieties and radical species to which molecules with nucleophilic groups such as amine- (R-NH<sub>2</sub>) and thiolate- (R-S<sup>-</sup>) spontaneously react with PD (details are explained in section 5.2).<sup>29,30</sup> Also, the PD coating is redox active, allowing electroless metallization and on surface synthesis of metal nanoparticles.<sup>31–33</sup> In contrast, polymers specially prepared to provide such functions are needed for LbL coatings.

Although the range of substrates amenable to PD and LbL methods have substantial overlap, it is generally accepted that PD is a more suitable method to modify a broader array of surfaces, including materials that are normally difficult to coat (e.g., low surface energy solids). For example, without adaptation of the basic approach, PD can easily functionalize most metal oxides and noble metals as well as low energy surfaces such as polytetrafluoroethylene, polydimethylsiloxane, polystyrene, poly(lactic-co-glycolic) acid, polycarbonate, poly-(caprolactone), graphene, carbon nanotubes. A more comprehensive list of materials functionalized by PD is summarized in Table 1. Notably, even extremely low energy surfaces including superhydrophobic/superomniphobic surfaces<sup>34</sup> can be modified with PD. Synergistic salt displacement at solid and liquid interfaces by catechol and amine groups<sup>11</sup> is one of important mechanisms why PD exhibits coating capability to such a broad spectrum of materials surfaces listed in Table 1. Also, PD layers utilizes a variety of multiple binding mechanisms such as catechol-metal coordinations, electrostatic interactions,  $\pi$ - $\pi$  interactions, hydrogen bonds, and covalent reactions (e.g., catechol-NH-R/catechol-S-R) depending upon chemistry of materials surfaces.<sup>16,35-40</sup> However, influences of solid substrates on PD thickness and homogeneity of coatings require further studies. In contrast to the LbL method, the PD method requires no significant surface preparation and aggressive cleaning of substrates prior to coating deposition. Finally, it should be emphasized that the PD and LbL coating methods in some cases may be complementary toolkits. For example, catechol or catecholamine moieties which are the building blocks of PD, have been chemically tethered or end-functionalized to polymers for use in LbL. Stability and substrate versatility are enhanced when catechol-conjugated polymers are used in LbL depositions.<sup>6</sup>

The two unique properties of a polydopamine coating, substrate flexibility combined with a variety of ad-layer properties by covalent/coordinate/noncovalent linkages with other molecules, allow virtually unlimited access to functional properties. The list of applications of PD is rapidly growing and seems to be limited only by the creativity of researchers using the method. A partial listing of PD applications demonstrated in just last 10 years include surfaces for stem and differentiated cell culture,<sup>85,131-134</sup> cell patterning,<sup>135-137</sup> microfluidics,<sup>138,139</sup> antimicrobial surfaces,<sup>46,140,141</sup> scaffold functionalization for tissue engineering,<sup>142-144</sup> bioimaging,<sup>145,146</sup> theragnostic,<sup>31,147,148</sup> photothermal therapy,<sup>149,150</sup> PLGA (nano)-particles<sup>66,151</sup> and capsules for drug delivery,<sup>125,129,130</sup> hydroxyapatite<sup>91,92,152</sup> or calcium carbonate surface mineralization,<sup>94,153,154</sup> artificial spores,<sup>155,156</sup> immobilization of photocatalysts and/or interplay between PD and photocatalysts,<sup>157-159</sup> Li-ion battery membranes,<sup>42,160-162</sup> Li-air battery electrolytes,<sup>163,164</sup> Li-sulfur battery cathode materials,<sup>165</sup> Zn-air cathode materials,<sup>166</sup> oil/water separation,<sup>167-169</sup> atomic transfer radical polymerization,<sup>77,170,171</sup> water detoxification,<sup>86,172</sup> carbonization,<sup>173-175</sup> membrane separation technologies,<sup>176-178</sup> organocatalysts,<sup>179,180</sup> and numerous others. It is certain that the scope of PD research and utilization will expand further in the years to come.

#### 4. PD COATING METHODS

Many of the advancements that have occurred during the first decade of PD research relate to modifications of the coating recipe that was originally published in 2007. In this section

we describe some of these key developments with an emphasis on choice of buffer and solvent, the use of chemical oxidants, utilization of external stimuli, and a description of a one-pot method that reduces the number of steps needed for functional coatings.

#### 4.1. Choice of Buffer and Solvent.

The original method of PD coating employed 2 mg/mL of dopamine hydrochloride (synonyms: 3-hydroxytyramine hydrochloride; 2-(3,4-dihydroxyphenyl)ethylamine) dissolved in Tris buffer, pH 8.0–8.5.<sup>1</sup> Dopamine concentration is an important tool in controlling deposition kinetics and roughness of surfaces. Recently, dopamine with a low concentration (<0.5 mg/mL) was used to functionalize nanostructures (i.e., particle,<sup>147,181</sup> tube,<sup>182</sup> and fiber<sup>68</sup>) because low concentration of dopamine could effectively reduce PD particle formation by self-polymerization and interparticle aggregation, and such aggregates inevitably increase roughness of PD coatings.<sup>181</sup> For instance, Au nanoparticles coated with dopamine (0.1 mg/mL) are stable as monodisperse nanoparticles, but small aggregates of particles are observed at 0.4 mg/mL dopamine concentration.<sup>181</sup> Likewise, a convenient method to minimize surface roughness is to decrease substrate immersion time to about 1–3 h in Tris buffer,<sup>183</sup> and the coating process can be repeated twice or three times if desired to control thickness.<sup>155</sup> Vincent Ball and co-workers clearly reveal concentration effects on the kinetics of PD deposition, thickness, roughness, and surface energy.<sup>184</sup> The maximal film thickness is increased linearly with dopamine concentrations from 0.1 to 5 mg/mL (i.e., 20 nm for 0.5 mg/mL, 25 nm for 1 mg/mL, and 25–40 nm for 2 mg/mL). In contrast, the thickness of PD varies at high concentrations of dopamine (3 and 5 mg/mL). In general, the thick PD films are rough compared with roughness of thin ones, and the surface energy of PD is independent with dopamine concentrations. In addition to concentrations, the maximal film thickness increases from pH 5 to 8.5 of dopamine solutions. However, during PD formation Tris<sup>17</sup> and unreacted dopamine<sup>20</sup> are unavoidably incorporated into the coating, which may alter the physicochemical properties of the coating and alter further chemical reactions. To avoid Tris buffer incorporation,<sup>185–187</sup> amine-free organic buffers (e.g., bicine) or inorganic (e.g., phosphate) buffers can be used instead, but the codeposition of PD nanoaggregates can be more problematic than when Tris buffer is used.

Choice of solvents is critical in some cases. While the vast majority of reports on PD describe the use of aqueous solvents, solvents with low surface tension such as methanol and ethanol can be used to modify hydrophobic and/or porous materials such as polyethylene (PE) membranes used in Li-ion batteries.<sup>42</sup> As we discuss further below, the use of organic solvents may be advantageous in other ways, for example enhancing the drying rate of treated substrates through fast evaporation, preventing degradation of hydrolyzable substrates, coimmobilization of water insoluble molecules, etc.

#### 4.2. Oxidation: Auto- Versus Chemical Oxidation.

Dissolved oxygen is essential for traditional PD formation via auto-oxidation at alkaline pH as was qualitatively shown in the original PD coating report.<sup>1</sup> Later, direct evidence regarding the importance of dissolved oxygen in aqueous solutions was demonstrated for the study of microwave accelerated PD coating (see section 4.3 for details).<sup>188</sup> Water-soluble, inorganic chemical oxidants such as sodium periodate (NaIO<sub>4</sub>), ammonium

per(oxodi)sulfate ((NH<sub>4</sub>)<sub>2</sub>S<sub>2</sub>O<sub>8</sub>), potassium per-manganate (KMnO<sub>4</sub>), copper sulfate (CuSO<sub>4</sub>), and Fe(III) have been widely used.<sup>12,57,189</sup> Vincent Ball and co-workers reported that superhydrophilic-superoleophobic PD coatings are achieved by the oxidants control.<sup>189</sup> In the presence of (NH<sub>4</sub>)<sub>2</sub>S<sub>2</sub>O<sub>8</sub> or CuSO<sub>4</sub> (above 10 mM oxidant concentrations), the PD formation results in heterogeneous coating on surfaces. However, the PD coating generated by NaIO<sub>4</sub> (the concentration lower than 30 mM) provides homogeneous surfaces. Furthermore, the thickness of the PD films in the presence of NaIO<sub>4</sub> was 65 nm after 1 h incubation, which was far higher than that of CuSO<sub>4</sub>-catalyzed PD films (43 nm). When CuSO<sub>4</sub> was used, copper ions (Cu<sup>2+</sup>) were also found in the PD coating, which is due to the chelation properties of the catechol moieties. Over the last several years, use of NaIO<sub>4</sub> in PD coating has become more widespread. Through optimization of pH, concentration of dopamine, and stoichiometric ratio of [NaIO<sub>4</sub>]/[dopamine], ultrafast and thick (>50 nm) PD coatings were obtained at room temperature.<sup>12</sup> Furthermore, hydrophilic coatings were obtained in the presence of large excess of NaIO<sub>4</sub> due to the formation of carboxylic acid groups on surfaces.

#### 4.3. Ultraviolet and Microwave Enhanced PD Coatings.

The generation of radical species by providing external energy such as ultraviolet (UV) light can also trigger PD formation as was shown by Levkin and co-workers.<sup>190,191</sup> The main advantages of using light are to control onset and termination of PD coatings, and to deposit patterns of PD on substrates. Furthermore, this light-induced method is effective from slightly acidic to basic pH ranges. Thus, when one utilizes acidic conditions and UV light, initiation and termination of PD deposition can easily be controlled. The use of UV irradiation in conjunction with chemical derivatives of dopamine provides an additional level of control. A. del Campo and co-workers used nitro-dopamine derivatives, which exhibit photocleavable properties with a leaving group of ortho-nitrophenyl ethyl moiety.<sup>192</sup> This chemistry provides new ways of controlling surface properties by detaching molecules that are tethered from surfaces.

Microwave irradiation of dopamine solution is another useful method to accelerate PD coating formation.<sup>188</sup> To achieve a PD coating thickness of 18 nm by the conventional alkaline PD coating method requires several hours, whereas microwave PD coating method takes only 15 min. The fast PD coating kinetics in the microwave technique is claimed to be due to enhancement of oxygen tension in the coating solution because of vibration-involved heating mechanisms. Interestingly, dissolved oxygen is ultimately removed in microwave heat-induced boiling, which was used to clearly demonstrate the importance of oxygen in PD formation.<sup>123</sup>

#### 4.4. One-Pot PD Coatings.

For surface tethering of molecules containing amine (–NH<sub>2</sub>) and thiol (–SH) groups to PD, the “one pot” method offers a simplified approach to forming PD coatings.<sup>143</sup> In the one-pot method, dopamine and polymer/biomolecule deposit simultaneously from solution, reducing the number of coating steps (Figure 4). An additional advantage of one-pot PD coating is that PD aggregation is largely suppressed because of dopamine/target molecule association. Representative one-pot coating studies include a comparative and quantitative analysis of

protein immobilization for the conventional two-step approach vs the one-pot method,<sup>193</sup> a tertiary amine coating for nanoscale silicification,<sup>160</sup> and creation of special wettability properties such as superomniphilic and omniphobic surface.<sup>34</sup> The initial study on one pot PD employed macromolecules included poly(vinyl alcohol), hyaluronic acid, dextran, and chitosan that have strong interactions with dopamine/PD,<sup>143</sup> although future studies will likely reveal numerous other polymers that can be employed with this method.

In general, we recommend use of a few milligrams of dopamine per mL of Tris pH 8.5 for 5–6 h in general purposes of solid substrate modifications. Overnight PD coating should be avoided for obtaining smooth surfaces because of generation of microsized PD aggregates. Also, a water/ethanol cosolvent recommends for PD coatings on hydrophobic surfaces or porous membranes because ethanol's low surface tension. For ad-layer formations with general purposes of molecular immobilizations, one-pot PD coating should be considered with the conventional two-step PD coatings to obtain high density molecular immobilizations.

## 5. TAILORING PD FUNCTIONALITY THROUGH BUILDING BLOCK DIVERSITY

One of the defining features of PD is undoubtedly the rich array of possibilities for tailoring surface properties for various applications. Here we review the two main approaches to functional versatility of PD coatings: the use of dopamine chemical derivatives in the primary deposition, and secondary ad-layer formation on an underlying PD “primer”.

### 5.1. Chemical Derivatives of Dopamine.

As shown in Figure 5, dopamine exhibits four possible sites (amine, alkyl and aromatic) for chemical derivatization (leaving aside O-substituted dopamine derivatives because they eliminate the catechol).<sup>194</sup> Perhaps the most obvious example of a dopamine derivative is 3,4-dihydroxy-L-phenylalanine (DOPA) ( $R^1 = \text{CO}_2\text{H}$ ), a biologically important free amino acid that is an intermediate in melanin formation and is decarboxylated to form dopamine in vivo. Interestingly, oxidative polymerization of DOPA to form polyDOPA coatings using a PD-like method is generally less successful than PD, possibly due to electrostatic repulsive interactions between neighboring carboxylic acid groups that may disrupt polymerization and/or aggregation of oligomeric polyDOPA subunits during coating formation.

Nevertheless, polyDOPA coatings were successfully applied to PE, PVDF, and PTFE membrane substrates and showed reduced static water-contact angles.<sup>195</sup> Subsequently, it was shown that most limitations of polyDOPA coating formation on noble metals, polymers, and oxides could be overcome through the use of high ionic strength deposition conditions.<sup>196</sup>

Norepinephrine ( $R^2 = \text{OH}$ ) has been widely used for coating formation, with the unique aspect being that conformational poly(norepinephrine) coatings are ultrasoft, with a uniform thickness  $\sim 20$  nm.<sup>197</sup> In contrast, under similar deposition conditions, the thickness of a PD coating ranges from 30–50 nm with a number of PD nanoparticles present. Norepinephrine-derived coatings have been formed on various materials including Si/SiO<sub>2</sub>, glass,



polystyrene, PDMS, and PTFE,<sup>198–201</sup> and on substrates of various morphologies such as microchips,<sup>202</sup> nanotubes,<sup>203</sup> nanoparticles,<sup>204</sup> nanofibers,<sup>205</sup> and sponges.<sup>206</sup> As an example of a benefit achieved through the use of chemical derivatives of dopamine, the additional hydroxyl group in norepinephrine allows for ring opening polymerization of lactone monomers from the surface in a two-step modification, resulting in grafted polyester.<sup>201</sup>

Substitutions at the primary amine ( $R^3$  position) have been the most widely used functional dopamine derivatives. Conjugations to the  $R^3$  primary amine intrinsically prevents indole formation, affecting the PD formation mechanism,<sup>194,207,208</sup> but surface modification may occur by catechol-to-catechol conjugation pathway. One functional group for modification at the  $R^3$  position is 2-bromoisobutryl bromide for initiating atom transfer radical polymerization (ATRP).<sup>209</sup> In addition, pyrrole, pyridine, and methacrylate as  $R^3$  substituents have been used in surface functionalizations.<sup>52,53,81,96,100,210–212</sup>

A representative  $R^4$  substituted dopamine is 6-nitrodopamine, which was mentioned previously as being photo-cleavable and used in the preparation of light-responsive smart surfaces.<sup>192</sup> This compound is primarily used to functionalize surfaces of inorganic nanoparticles such as iron oxide and titania.<sup>213–217</sup> 6-Nitrodopamine is rather oxidation resistant compared to dopamine due to the presence of the electro-negative nitro groups. This potentially results in maintaining strong binding affinity to nanoparticles.<sup>215</sup> Another  $R^4$ -substituted dopamine is 5-hydroxydopamine<sup>218</sup> (we describe the use of amine-free gallol-derived compounds below in Section 6.2).

## 5.2. Secondary (ad-layer) Functionality.

The second approach for tailoring functionality is to utilize the intrinsic chemical reactivity of the surface of PD to deposit an ad-layer. These secondary reactions may exploit noncovalent binding interactions, or covalent reactions with molecules containing nucleophilic or other reactive groups. Nearly all proteins, peptides, end-functionalized oligonucleotides, and a large population of small molecules are amenable to this approach. Other molecules, including synthetic polymers, can be modified or synthesized with functional groups enabling reactions with PD. Reaction conditions for ad-layer grafting are generally the same as PD formation (buffers with basic pH).

Early studies in this area involved thiol- or amine-terminated polymers grafted onto PD-coated surfaces through thiolcatechol or amine-catechol adducts by Michael-type addition reactions and/or Schiff-base formations (Figure 6),<sup>1,38–40,46,77,176,219–225</sup> with subsequent expansion to bio-molecules (peptides, protein enzymes and oligonucleotides).<sup>226,227</sup>

Examples of proteins and peptides successfully immobilized onto PD are numerous and include albumin,<sup>228,229</sup> lysozyme,<sup>230</sup> bone morphogenic protein-2,<sup>142,231–233</sup> trypsin,<sup>234</sup> alkaline phosphatase,<sup>193</sup> antifreeze proteins,<sup>235</sup> collagen,<sup>236</sup> collagenase,<sup>230</sup> aquaporin,<sup>237</sup> vitronectin (VN2)-derived peptides,<sup>238</sup> Arg-Gly-Asp (RGD) peptides,<sup>142</sup> epidermal growth factor,<sup>239</sup> and many others. Ad-layer immobilization of (strept)avidin provides even more versatility through strong avidin–biotin reversible interactions.<sup>229</sup>

In the case of biomolecules, the promiscuous reactivity of catechols and quinones at the PD surface is normally thought to make these reactions poorly specific at best, however a recent report of a chemoselective reaction between catechol and the N-terminal amine of proteins and peptides suggests opportunities for chemospecific conjugations to PD.<sup>240</sup> An interesting recent publication described the surprising orientation-specific immobilization of antifreeze proteins onto PD surfaces.<sup>235</sup> Apparently the amino acid composition exposed on the ice binding and nonice binding faces of the protein allowed for selective immobilization in such a way as to orient the protein with the nonice binding oriented away from the surface, reducing ice formation at the modified surface.<sup>235</sup>

PD is also a good platform for surface tethering and release of small molecule drugs and therapeutic RNAs<sup>66,130,241,242</sup> through electrostatic interactions, hydrogen bonds,  $\pi$ - $\pi$  stacking, cation- $\pi$  interactions.<sup>103,141,243-245</sup> For instance, the amount of bound siRNA on PD substrates is larger than that of unmodified substrates and shows sustained release profiles over at least 7 days.<sup>242</sup> In another study, siRNAs were successfully loaded onto surfaces of manganese oxide nanoparticles for delivery to target cells.<sup>246</sup>

## 6. FUTURE OPPORTUNITIES AND CHALLENGES IN PD COATINGS

With a view toward guiding further development of PD and related coating technologies in the coming years, in the section below we describe several areas that we feel are deserving of more attention by researchers.

### 6.1. Improving the Mechanical Properties of PD.

Considering its chemical resemblance to mussel adhesive proteins and its ability to form conformal coatings on a multitude of solid surfaces, one would predict that PD coatings should function well as mechanically robust coatings. An application of PD in which mechanical properties are important is in fiber and particulate composites, where dopamine has been used to enhance wetting and adhesion between phases (recently reviewed by Ball<sup>247</sup>). As an example of PD applications in composites, the mechanical properties of bioceramics composed of hydroxyapatite and gelatin modified with silane (HAp-Gemosil) was improved by incorporation of PD.<sup>248</sup> The compressive strength of PD-incorporated HAp-Gemosil (HAp-Gemosilamine) is approximately 100 MPa, higher than that of HAp-Gemosil (~80 MPa). Several other recent reports have been published showing the ability of PD to enhance mechanical properties of composites.<sup>249,250</sup>

The application of PD to mechanical composites notwithstanding, the anecdotal experience of our laboratories, and that of several others, is that PD coatings in their current form do not perform particularly well as mechanical adhesives or in a context that requires resistance to delamination or abrasion. This is particularly true on flat surfaces and when PD is applied to low-surface-energy materials. Widespread incorporation of PD into products or components of devices may be further hindered unless improvements in PD mechanical properties can be realized.

Achieving these goals will likely require better understandings of chemical composition and physical properties of PD coatings, which might lead to develop next-generation PD coating

with improved mechanical properties. Surprisingly, few investigations of mechanical properties of PD have been performed in the past.<sup>123,245,251–254</sup> The elastic modulus of PD has been measured to be in the GPa range in the dry state,<sup>123, 251</sup> suggesting that PD is quite rigid. The importance of primary amines in PD adhesion was measured using the surface forces apparatus, suggesting that a role for surface salt displacement as well as  $\pi$ – $\pi$  and cation– $\pi$  interactions in adhesion.<sup>245</sup> Moreover, chemical cross-linking of amines in the coating offers a mechanism for increasing PD modulus.<sup>251</sup> You and co-workers performed an investigation of lap shear mechanical adhesion of PD films, characterizing the mechanical adhesion of PD while at the same time illustrating how chemical analogs affect adhesion strength.<sup>252</sup> In addition to providing insight into PD polymerization mechanisms, an interesting outcome of this study was that extension of the alkyl chain linker between catechol and amine did not affect adhesion strength. New methods of measuring the adhesion strength of PD films to substrates may prove useful in screening new PD formulations for improved adhesion to substrates.<sup>254</sup>

## 6.2. Expanding the Chemical and Biological Diversity of PD-like Coatings.

A recent development in the field involves the use of nitrogen-free phenols and polyphenols as coating precursors. Catechol-containing molecules that form coatings include hydrocaffeic acid,<sup>255,256</sup> alkylcatechol,<sup>257–262</sup> and thiol-terminated catecholic monomers.<sup>263</sup> Gallol (2,3,4-trihydroxyphenyl) based molecules such as pyrogallol and tannic acid are also emerging as useful precursors for coating formation. Surface coatings based on metal phenolic networks (MPN) are well-known examples.<sup>264,265</sup> In the case of MPN coatings based on tannic acid, addition of transition metals such as Fe(III), Cr(III), V(III), Zn(II), or Cd(II)<sup>266</sup> to tannic acid exhibits material-independent surface coatings by metal coordination network formation. Because of the fast metal–ligand coordination kinetics, the coating forms rapidly, which is an advantage of MPN. In contrast, PD coating requires several hours unless special fast coating processes are used. MPN coatings formed in this manner can provide support for surface PEGylation.<sup>267</sup> Other important applications of MPN coatings formed in this manner can provide support for surface PEGylation.<sup>267</sup> Other important applications of MPN coatings include protective coating for cell attachment,<sup>268,269</sup> tooth desensitization,<sup>270</sup> heavy metal removal,<sup>271</sup> and protein immobilization.<sup>272,273</sup>

Nitrogen-free catechol and gallol-containing precursors are capable of forming coatings in the absence of metals via auto-oxidation in a manner that is similar to PD formation. The observation of spontaneous adsorption of phenolic compounds from beverages (e.g., tea, wine) rich in plant polyphenols onto surfaces led to a significant expansion of potential building blocks for spontaneous coating formation.<sup>274</sup> This approach was first demonstrated with pyrogallol, tannic acid, epigallocatechin gallate (EGCG), epicatechin gallate (ECG), and epigallocatechin (EGC), which form nanoscale coatings in a kinetically controlled auto-oxidation process that resembles that of PD formation, and then later expanded to at least 15 natural compounds that are known to form coatings by auto-oxidation.<sup>275</sup> Many of the inherent advantages of PD, namely simplicity, substrate versatility and multifunctionality, are shared by plant polyphenol derived coatings. Some of these coatings have the advantage of being colorless, unlike PD coatings.<sup>274</sup> Furthermore, some of the biological properties of plant polyphenols such as antibacterial activity, antioxidant activity, and other properties are

conferred into the coatings. Already there is a rapidly growing body of reports employing nitrogen free polyphenol coatings for a variety of purposes.<sup>267,273,276–304</sup> An important consequence of the enormous biological diversity of plant polyphenols is that hundreds if not thousands of natural plant polyphenols can now be considered as coating precursors, with a broad range of biological and chemical properties. We anticipate that this large biological toolbox of coating precursors will be increasingly exploited in the coming years, leading to unforeseen coating properties and applications.

## 7. CONCLUSIONS

In its first ten years, PD has proven to be one of the most powerful and widespread surface coating methods due to its material-independent coating ability, the simplicity of the coating deposition process, and the unique and broad ranging capabilities for ad-layer formation leading to numerous practical uses. The thickness and PD coating properties can be tailored by parameters such as coating time, pH, solvent, dopamine concentration, and chemical additives including metal ions. In addition, target molecules such as natural/synthetic polymers, proteins, peptides, oligonucleotides, and numerous small molecules (including drugs) can be readily immobilized by ad-layer formation or through one-pot coating methods. The use of chemical analogs of dopamine promises to further expand the properties and applications of PD coatings. Important challenges facing the field in the future include better understanding of PD formation mechanisms and elucidation of the chemical structure of PD, enhancement of mechanical robustness of PD, and extension of the general PD coating approach to more chemically diverse building blocks such as the nitrogen-free catechol and gallol compounds found in plant tissues. With advancements in these areas, it is likely that the family of PD and related coatings will be widely implemented in biomedical, energy, consumer products, agricultural, military, and other sectors.

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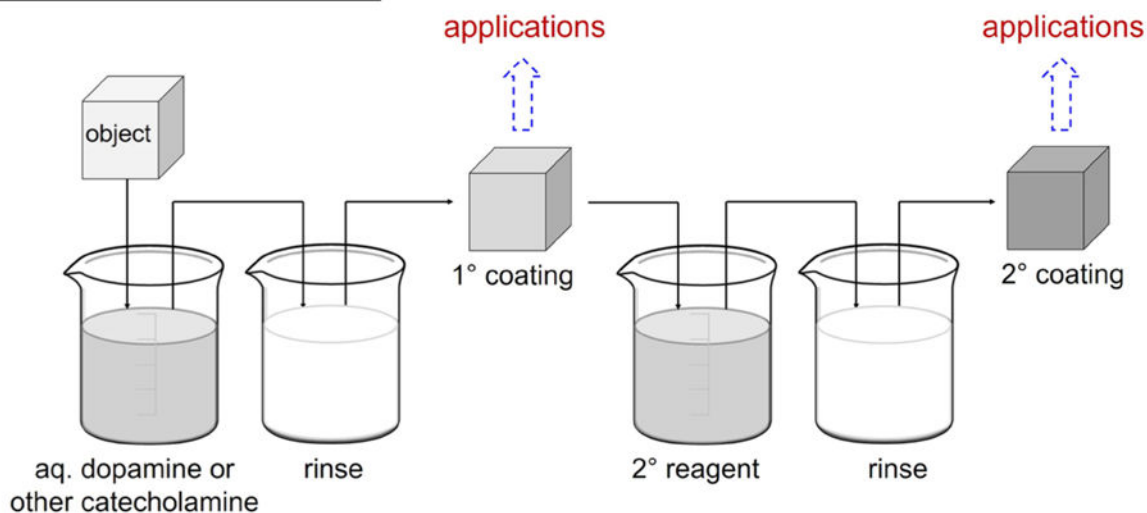
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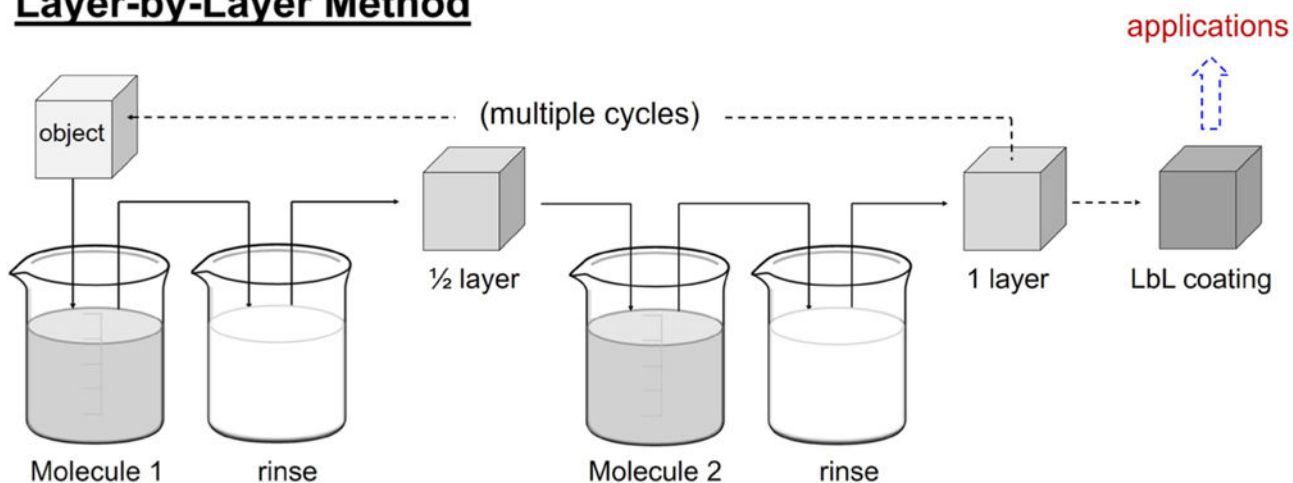
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## Polydopamine Method

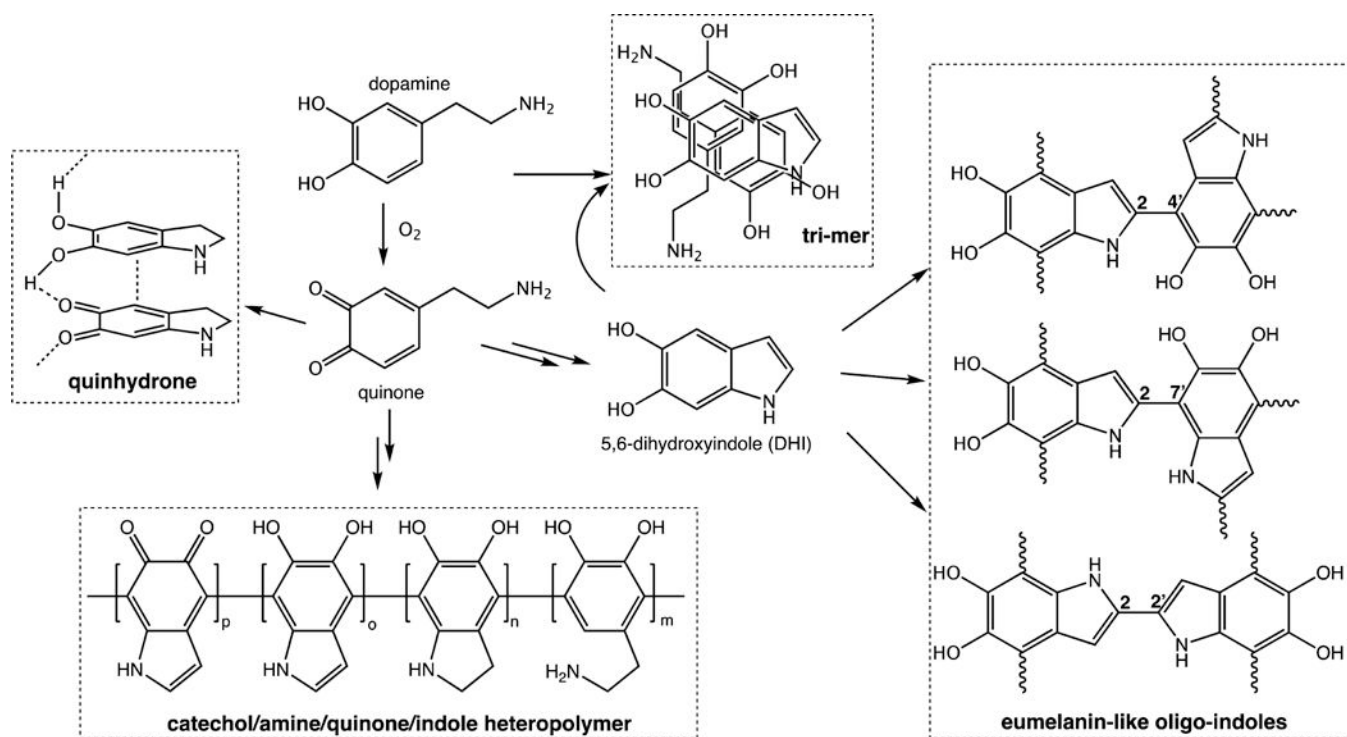


## Layer-by-Layer Method

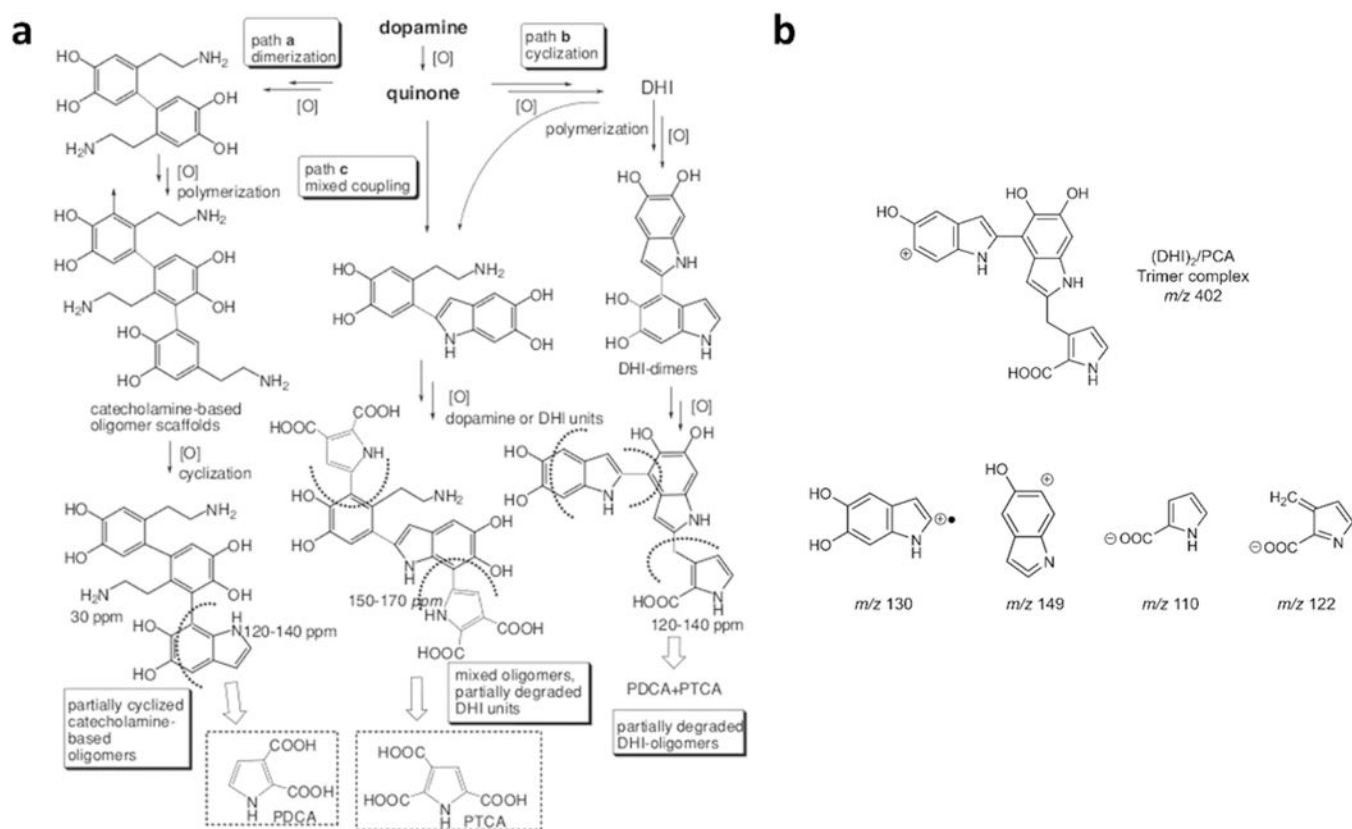


**Figure 1.**

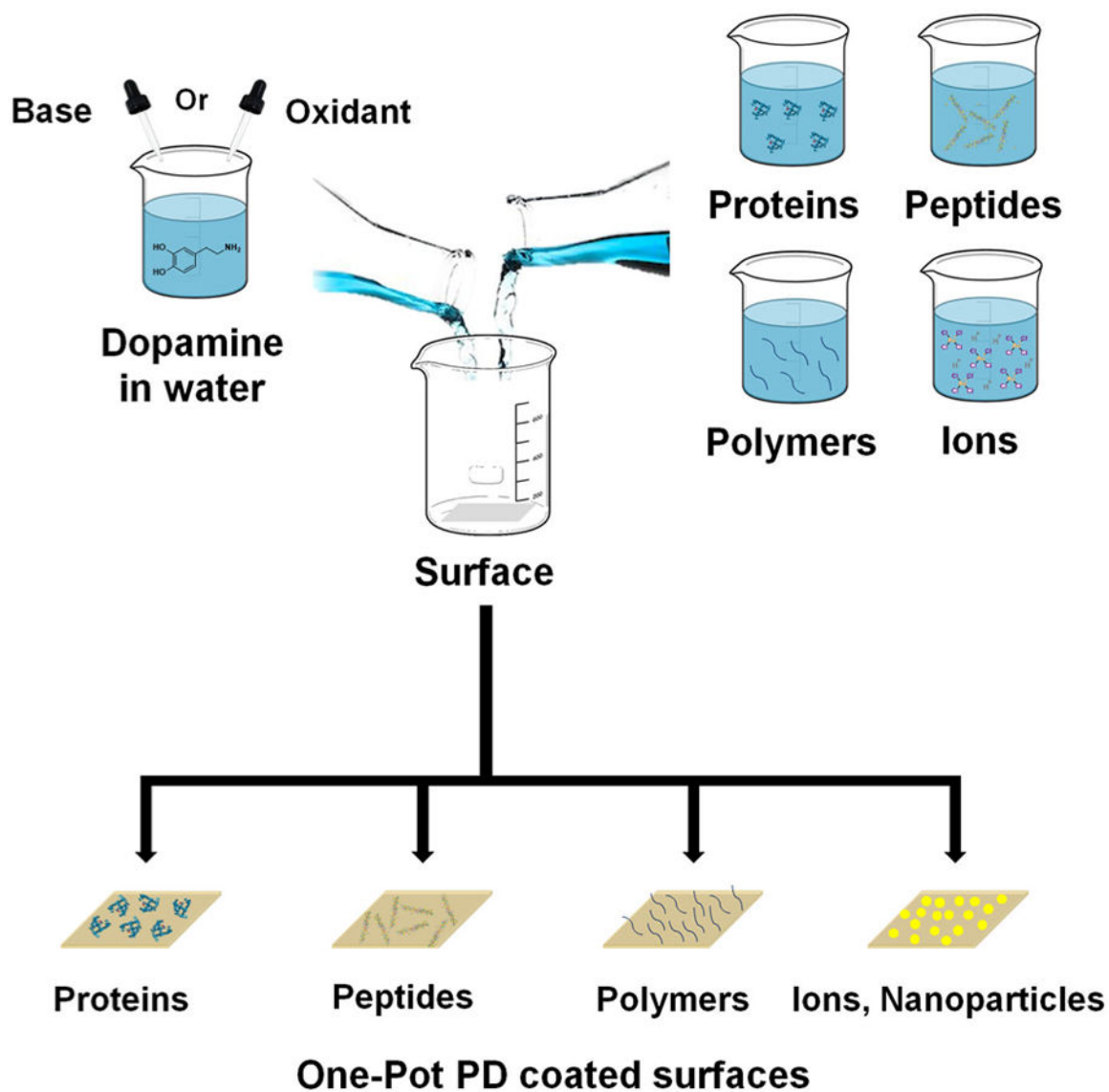
Schematic illustration of PD coating method with comparison to LbL coatings. Top: the traditional PD method takes place spontaneously in alkaline aqueous solutions, or with the addition of oxidants. A subsequent ad-layer (secondary coating) step can be undertaken. Secondary reagents are usually amine or sulfhydryl containing nucleophiles such as peptides, proteins, oligonucleotides, or nucleophilic natural or synthetic polymers. Bottom: the LbL method involves cyclic adsorption of typically polymeric components with intermediate rinsing steps. Often, dozens or even hundreds of cycles are used to build the coating.



**Figure 2.** Current theories of polydopamine structure and formation. Auto-oxidation of dopamine leads to the formation of dopamine-quinone and 5,6-dihydroxyindole. Proposed mechanisms for polydopamine formation range from noncovalent self-assembly of subunits to form quinhydrone or trimer assemblies, and covalent coupling of subunits to yield a catecholamine/quinone/indole heteropolymer or eumelanin-like oligo-indoles. Adapted with permission from refs 13, 15, and 20. Copyright 2013 American Chemical Society, 2014 American Chemical Society, and 2012 Wiley-VCH.



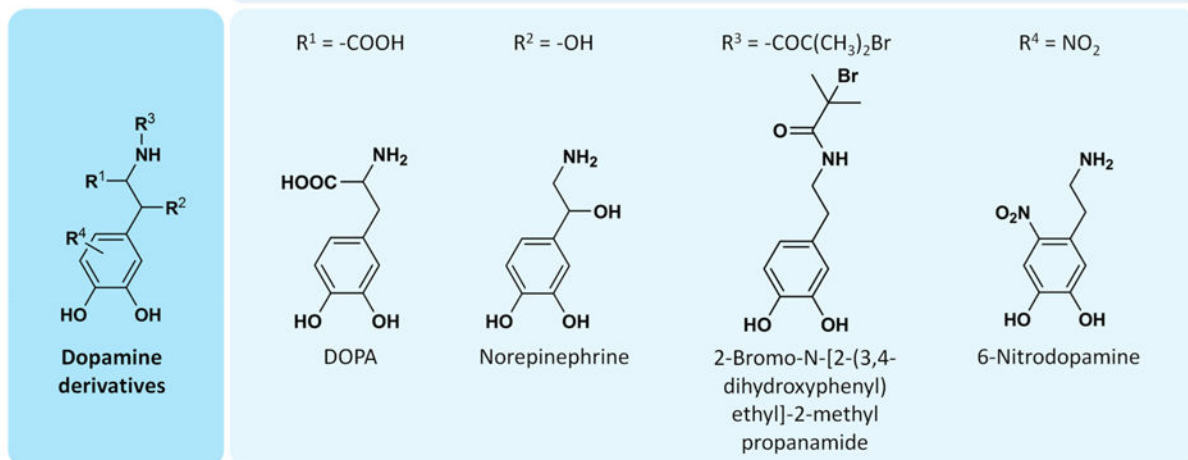
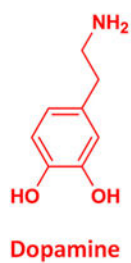
**Figure 3.** Pathway of pyrrolecarboxylic acid-involved PD formation. (a) Analysis of oxidative degradation products suggests PD formation pathways. Pyrrole-2,3-dicarboxylic acid (PDCA) is originated from partially cyclized catecholamine oligomers. Pyrrole-2,3,5-tricarboxylic acid (PTCA) is an outcome of DHI unit degradation. Reprinted with ref 17. Copyright 2013 Wiley-VCH. (b) DHI<sub>2</sub>/PCA trimer complex as a building block of PD. Reprinted with permission from ref 18. Copyright 2014 American Chemical Society.



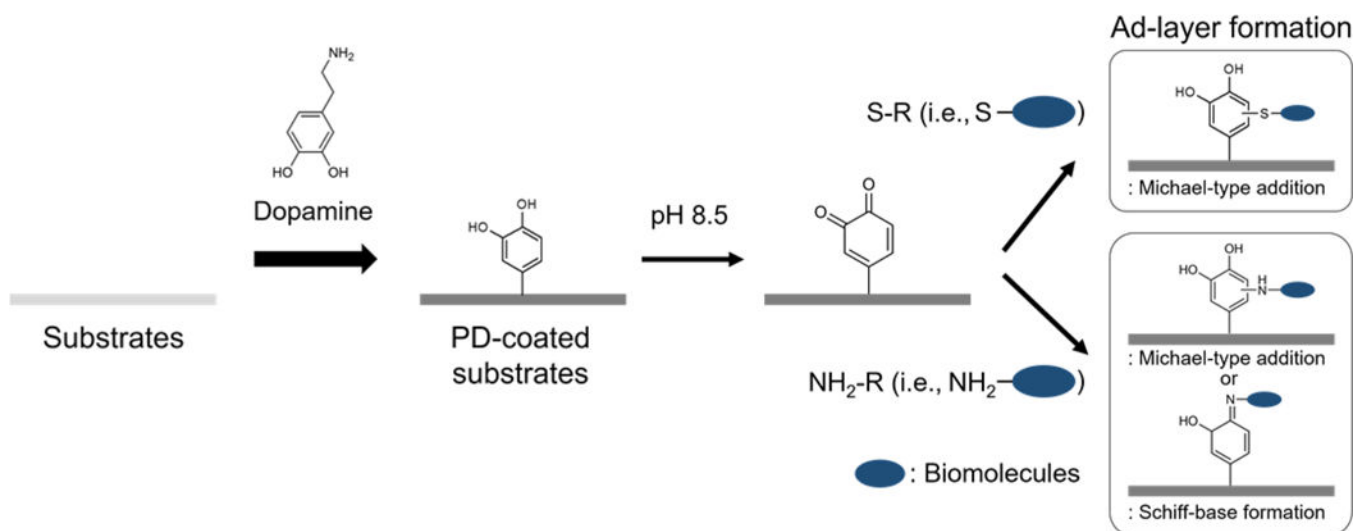
**Figure 4.**

One-pot PD coatings. The one-pot method for preparing PD coatings utilizes a precursor solution containing a mixture of dopamine and molecules to be coimmobilized with PD. The method can use either the auto-oxidation approach at basic pH solution or chemical oxidants to produce functional substrates.





**Figure 5.** Chemical structures of dopamine derivatives. Opportunities exist for chemical substitutions of dopamine at alkyl ( $R^1$ ,  $R^2$ ), amino ( $R^3$ ), and aromatic ( $R^4$ ) sites, offering the potential for tailoring the formation and physicochemical properties of PD coatings.



**Figure 6.** Ad-layer functionalization with thiol- and amine-containing biomolecules on PD-coated substrates. Adapted with permission from refs 38, 39, and 40. Copyright 1999 American Chemical Society, 1987 American Society for Biochemistry and Molecular Biology, and 2006 American Chemical Society.

**Table 1.**

## List of Substrates Successfully Coated with PD

materials (substrates)	substrate form	ref
polystyrene (PS)		1
	nanofiber	41
polyethylene (PE)		1
	membrane	42
polypropylene (PP)		43, 44
	nanofiber	45
Polycarbonate (PC)		1, 46
polyethylene terephthalate (PET)		1, 47
	PET/Ag hybrid fiber	48
polyester		44
poly(dimethylsiloxane) (PDMS)		1, 47
	PET/Ag hybrid fiber	48
polyester		44
poly(dimethylsiloxane) (PDMS)		1, 49–53
polytetrafluoroethylene (PTFE, Teflon)		54, 55
	microtube	56
poly(ether sulfone)	membrane	57
polyvinyl alcohol (PVA)	nanofiber	58
PVA/polyacrylic acid (PAA)	nanofiber	59
poly(vinylidene fluoride)	nanofiber	60
poly(vinylidene fluoride) (PVDF)	membrane	61
polyether ether ketone (PEEK)		1, 62
	membrane	63
polyurethane (PU)		1
	sponge/foam	64, 65
poly(lactic-co-glycolic acid) (PLGA)	nanoparticle	66
poly(ε-caprolactone) (PCL)	fiber	67
	particle	68
	scaffold	69, 70
polyimide (PI)		55
cellulose	membrane	71
	filter paper	72
paper		73
silk	fiber	74
nylon	membrane	57
graphene		54

materials (substrates)	substrate form	ref
graphene oxide (GO)		75, 76
carbon nanotube (CNT)		77, 78
diamond		79
diamond-like carbon		80
SiO <sub>2</sub>		1, 81
	nanoparticle	82
	membrane	83
	porous scaffold	84
Si <sub>3</sub> N <sub>4</sub>		1
glass		1, 85
	bead	86
tetraethyl orthosilicate	nanofiber (sol-gel)	87
clay		88
quartz		1, 89
fertilizer		90
mica		55
hydroxyapatite		1
	crystallization	91, 92
calcium phosphate	cement	93
calcium carbonate	powder	94
TiO <sub>2</sub>		1, 95, 96
	nanoparticle	97
	nanowire	98, 99
	nanotube	100, 101
ZrO <sub>2</sub>	nanocomposite	102
Nb <sub>2</sub> O <sub>3</sub>		1
Fe		55
Fe <sub>3</sub> O <sub>4</sub>	nanoparticle	103
Pd		1
	nanoparticle	104, 105
Pt		1, 106, 107
Cu		1, 108, 109
Ag		1
	nanostructure	110
Au		1, 54, 111
ZnO <sub>2</sub>	nanorod	112
Al		44
Al <sub>2</sub> O <sub>3</sub>	nanoparticle	113
Al(OH) <sub>3</sub>	particle	114

materials (substrates)	substrate form	ref
GaAs		1
In <sub>2</sub> O <sub>3</sub> /SnO <sub>2</sub> (Indium Tin Oxide, ITO)		115
stainless steel		1, 116
	porous	117
CdS/CdSe	quantum dot	118
virus, <i>E.coli</i>		119, 120
superhydrophobic surface		121
water surface	air/water interface	122–124
PD capsule		125–130

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