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REVIEW

Polydopamine-Based Biomaterials in Orthopedic Therapeutics: Properties, Applications, and Future Perspectives

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Abstract: Polydopamine is a versatile and modifiable polymer, known for its excellent biocompatibility and adhesiveness. It can also be engineered into a variety of nanoparticles and biomaterials for drug delivery, functional modification, making it an excellent choice to enhance the prevention and treatment of orthopedic diseases. Currently, the application of polydopamine biomaterials in orthopedic disease prevention and treatment is in its early stages, despite some initial achievements. This article aims to review these applications to encourage further development of polydopamine for orthopedic therapeutic needs. We detail the properties of polydopamine and its biomaterial types, highlighting its superior performance in functional modification on nanoparticles and materials. Additionally, we also explore the challenges and future prospects in developing optimal polydopamine biomaterials for clinical use in orthopedic disease prevention and treatment.

Keywords: polydopamine, biomaterials, nanoparticles, bone, orthopaedics

Introduction

Polydopamine (PDA), a polymer synthesized from dopamine monomers, is a key component found in human melanin. It has demonstrated that PDA exhibits exceptional adhesiveness, numerous covalently modifiable functional groups, significant near-infrared (NIR) absorption capacity,[1] and favorable biodegradability. Due to these advantages, PDA is commonly used for surface modification of materials, attracting interest from the scientific community. PDA has been thoroughly investigated across a spectrum of biomedical fields, including cell culture, drug delivery, biomaterial coatings, biomedical imaging, tissue engineering, and biosensing applications.[2,3] PDA has emerged as a versatile biomaterial with immense potential in orthopedic therapeutics. Studies have demonstrated that PDA nanoparticles exhibit effective performance in the delivery and sustained release of anti-osteoarthritis (OA) drugs. Additionally, applying PDA nanoparticle coatings to biomaterial surfaces enhances their adhesiveness and antibacterial properties,[4] while PDA's antioxidative properties provide protection to biomaterials within tissues. Research on bone reconstruction and osteogenic differentiation has shown that PDA nanoparticles are capable of downregulating pro-inflammatory factors in chondrocytes, creating a favorable microenvironment, regulating cell behavior, and promoting the vitality, adhesion,

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migration, and osteogenic differentiation of mesenchymal stem cells (MSCs).[5] Polydopamine-based biomaterials, despite being at a rapid development stage, offer promising avenues for the prevention and treatment of orthopedic diseases. This review aims to comprehensively explore the current state of polydopamine-based orthopedic therapeutics, highlighting its properties, applications, achievements, challenges, and future directions.

Synthesis and Properties of Polydopamine: Methods and Applications

PDA forms from dopamine (DA) under alkaline conditions through oxidation, cyclization, and rearrangement to form indolequinone, followed by further reactions with DA molecules,[6] as illustrated in Figure 1. The polymerization of PDA is typically achieved through three methods, including solution oxidation, enzyme oxidation, or electro-polymerization.[7] Among these methods, solution oxidation remains the most widely used due to its simplicity. Although this method functions under mild conditions and does not necessitate complex equipment, it exhibits sensitivity to various factors, including DA concentration,[8] oxidant content, temperature, pH, and stirring speed. PDA is usually polymerized at room temperature (20–25°C). Although higher temperatures, up to 60°C, can accelerate dopamine polymerization, they may also lead to non-specific aggregation.[9] Oxygen plays a vital role and is sufficient for PDA polymerization. The addition of ozone accelerates the dopamine polymerization process within the pH range of 4.0 to 10.0 and extends the temperature range, allowing polymerization even at low temperatures, such as 5°C.[10] The optimal pH range for dopamine (DA) polymerization is between 8.5 and 9.5. Previous study revealed that DA molecules first undergo a pH-induced auto-oxidation reaction to form dopamine quinone (DAQ).[11] DAQ molecules then crosslink through the formation of biphenyl bonds, leading to the formation of polycatecholamine intermediates, which further grow into the PDA film. During the polymerization of PDA, π-π interactions and hydrogen bonding play crucial roles. Dopamine (DA) oxidizes to quinone, which can cyclize into 5,6-dihydroxyindole (DHI). These three monomers can coexist in solution, each contributing to stacking and polymerization. Between monomer layers, π-π stacking and hydrogen bonding occur, while covalent bonds form between monomers at multiple sites.[11] Monomers bond through benzene rings to form lengthy polymer backbone chains, with the PDA backbone containing multiple types of constructing monomers. Indole groups may crosslink, lengthening the side chain or stretching the backbone. Studies show that two cross-plane dopamine monomers physically interact during polymerization, clasping one DHI and generating a physical trimer.[12,13] DA forms hydrogen bonds with molecules and other functional groups, contributing to its high adhesion strength to both organic and inorganic surfaces.[14–16] In conclusion, π-π interactions and hydrogen bonding are fundamental to the properties and applications of PDA materials, significantly influencing their structural integrity and adhesion capabilities.

On the other hand, the laccase-catalyzed method is suitable for alkaline-sensitive materials, as it allows PDA polymerization to be completed at a pH of 5.5[17] Usually, the polymerization reaction can take several hours or even a day.[18] During the polymerization process, dopamine molecules gradually polymerize and deposit on surfaces or form nanoparticles. Ultrasound can increase the deposition rate of PDA coatings as polymer films.[19] Additionally, adding a Fe catalyst can accelerate PDA polymerization at pH 7.0 and even facilitate it at lower pH levels down to 4.0.[20] Typically, the rate of PDA polymerization of this method is notably slow, and the resulting PDA often settles at the bottom of the container, posing challenges for researchers. Enzyme oxidation, while more complex than solution oxidation, is considered relatively environmentally friendly and typically involves enzymes such as tyrosinase, polyphenol oxidase (also known as laccase), and urase.[21] Electro-polymerization can produce PDA more rapidly than solution/enzyme

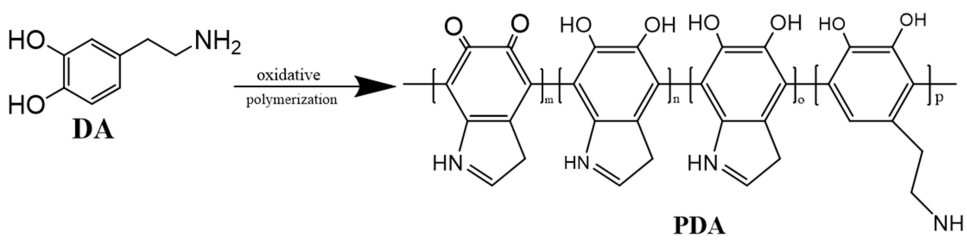

Figure 1 The Self-Polymerization Process of Polydopamine.

oxidation and avoids employing environmentally unfriendly chemical oxidants. This method can be readily applied to various metal substrates, rendering PDA suitable for surface modification of metal implants.[22] Structurally, PDA resembles the adhesive proteins secreted by invertebrate mussels (3, 4-dihydroxyphenylalanine). It is broadly recognized that multiple functional groups in PDA, comprising catechol groups, amino groups, carboxyl groups, indole units, and quinone functionalities, play a key role in its multifunctionality, resembling the adhesive properties of mussels. The abundance of primary and secondary amines and ortho-quinones in PDA's structure is instrumental in its strong adhesion, enabling it to adhere to nearly any material surface. The thickness and stability of the PDA adhesive film depend on the concentration of monomeric dopamine during self-polymerization. Prior research indicated that the elongation of the alkyl chain connecting the ortho-quinone and amine groups in PDA does not impact its adhesion strength.[23] Primary amines of PDA can be readily covalently modified, its excellent modifiability allows for tailored designs to suit specific therapeutic needs. Consequently, PDA represents an ideal adhesive material capable of modifying nearly all material surfaces and offers biocompatibility, presenting significant safety advantages.

Polydopamine Biomaterials: Properties and Types

PDA is derived from the polymerization of dopamine (DA), a critical neurotransmitter exist in the human body. DA plays a significant role in motor control, emotion regulation, endocrine regulation, cognitive function, and reward pathways. Consequently, polydopamine (PDA) degrades into DA, which contributes to its excellent biocompatibility and low cytotoxicity. Furthermore, PDA does not provoke significant immune responses, making it ideal for implant fabrication and drug delivery systems. Inspired by the adhesive proteins found in mussels, PDA exhibits strong adhesive properties. It contains a high density of primary amines, secondary amines, and catechol groups, which allow it to adhere to nearly any material surface. This versatility makes PDA suitable for coating a wide variety of substrates. PDA contains numerous functional groups that facilitate chemical modifications. This allows for the easy conjugation of various molecules, including drugs, proteins, and other biomolecules, enabling PDA to serve as a versatile platform for targeted drug delivery and other therapeutic applications. PDA's unique molecular structure and specific functional groups enable it to absorb light and convert it into heat, imparting special photothermal properties. The catechol groups in PDA contain aromatic rings with hydroxyl groups that can absorb light, particularly in the near-infrared (NIR) region. When exposed to NIR light, these groups undergo electronic transitions, generating heat. The π-conjugated system within the PDA polymer further enhances light absorption across a wide range of wavelengths, including the NIR region, allowing for efficient conversion of absorbed light into thermal energy. This makes PDA suitable for photothermal therapy applications.[24–26] Polydopamine can be engineered into various biomaterial forms, including nanospheres, nanocapsules, nanofilms, and nanocomposites. Some representative electron microscope images of the PDA biomaterials are shown in Figure 2, including PDA porous particles, PDA/Cu_2O coating, Ag/TiO_2/PDA-bamboo, and PU-PDA, among others. These forms offer distinct advantages in terms of drug loading and release kinetics, providing precise control over therapeutic interventions. PDA has been successfully applied in coating or modifying various nanostructures, enhancing their specific surface areas and increasing the surface available for cell and biomolecular interactions. Based on existing studies, PDA nanostructures can be classified into several categories, as illustrated in Figure 3. These include: (1) organic PDA nanoparticles formed by self-oxidation and polymerization in alkaline solutions without templates; (2) hollow PDA nanoparticles, nanocapsules, nanotubes (NTs), and nanorods, which result from deposition on movable templates; and (3) PDA nanoparticles co-assembled with other biologically active components, such as PDA core/shell nanoparticles and PDA nanofilms that adhere to the surface of other nanostructures.[25,27,28] During the preparation and application of PDA nanoparticles (PDA NPs), an increase in the pH of the alkali solution may reduce the diameter of the nanoparticles.[29]

Application of Polydopamine Biomaterials in Orthopedic Disease Prevention and Treatment

Currently, extensive research has led to the development of various organic and inorganic nano-drug delivery systems, including silica nanoparticles, titanium nanotubes, gold nanoparticles, calcium phosphate nanoparticles, chitosan nanoparticles, liposomal nanoparticles, and polymer nanoparticles, along with micelles, and dendritic macromolecules, aimed

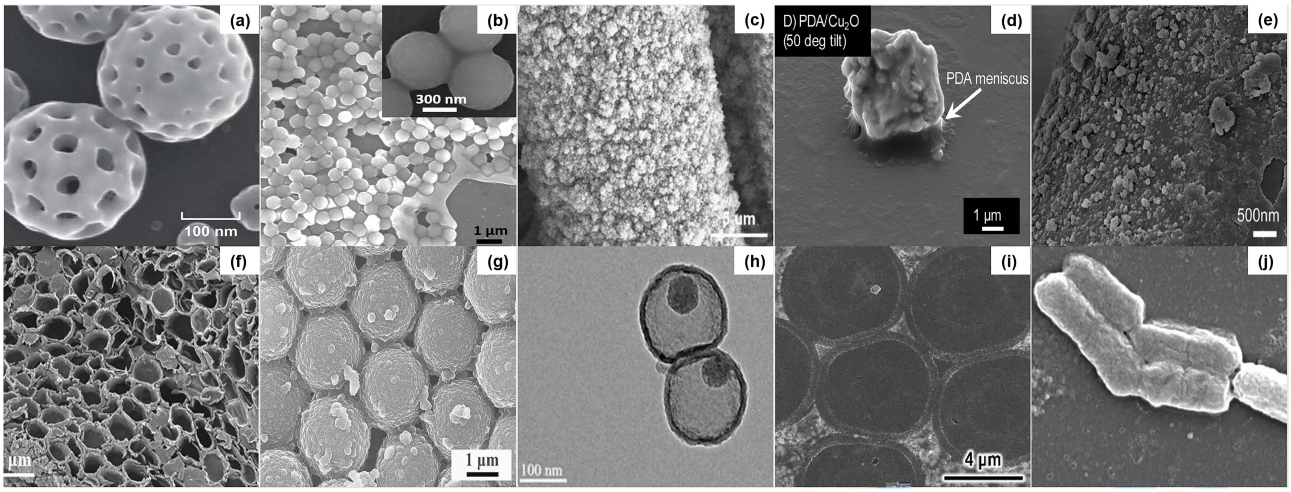

Figure 2 Overview of different SEM structures of polydopamine materials. (**a**) SEM images of the PDA porous particles. Adapted from Mei S, Kochovski Z, Roa R, et al. Enhanced catalytic activity of Gold@Polydopamine nanoreactors with multi-compartment structure under NIR irradiation. *Nanomicro Lett*. 2019;11(1):83. Creative Commons.[30] (**b**) SEM images of the PDA@SiO_2. Adapted from Khan MZH, Daizy M, Tarafder C, Liu X. Au-PDA@SiO(2) core-shell nanospheres decorated rGO modified electrode for electrochemical sensing of cefotaxime. *Sci Rep*. 2019;9(1):19041. Creative Commons.[31] (**c**) SEM images of cotton/pDA/MnO_2. Adapted with permission from Zhang Y, Zhao Z, Li D, et al. In situ growth of MnO2 on pDA-templated cotton fabric for degradation of formaldehyde. *Cellulose*. 2022;29(13):7353–7363.[32] (**d**) SEM images of the PDA/Cu_2O coating. Adapted from Behzadinasab S, Williams MD, Hosseini M, et al. Transparent and sprayable surface coatings that kill drug-resistant bacteria within minutes and inactivate SARS-CoV-2 virus. ACS Appl Mater Interfaces. 2021;13(46):54706–54714. Copyright © 2021 American Chemical Society.[33] (**e**) SEM images of the non-woven polydopamine (NW@PDA) fabrics. Adapted from Zhang Z, Si T, Liu J, Zhou G. In-situ grown silver nanoparticles on nonwoven fabrics based on mussel-inspired polydopamine for highly sensitive SERS carbaryl pesticides detection. *Nanomaterials*. 2019;9(3). Creative Commons.[34] (**f**) SEM images of the Ag/TiO_2/PDA-bamboo surface. Adapted from Liu G, Lu Z, Zhu X, et al. Facile in-situ growth of Ag/TiO(2) nanoparticles on polydopamine modified bamboo with excellent mildew-proofing. *Sci Rep*. 2019;9 (1):16496. Creative Commons.[35] (**g**) SPS-MS with PDA-PPy coatings. Adapted from Xie C, Li P, Han L, et al. Electroresponsive and cell-affinitive polydopamine/polypyrrole composite microcapsules with a dual-function of on-demand drug delivery and cell stimulation for electrical therapy. *NPG Asia Materials*. 2017;9(3):e358–e358. Creative Commons.[36] (**h**) TEM images of MSN@PDA. Adapted from Tran HQ, Bhave M, Xu G, Sun C, Yu A. Synthesis of polydopamine hollow capsules via a polydopamine mediated silica water dissolution process and its application for enzyme encapsulation. Front Chem. 2019;7:468. Creative Commons.[37] (**i**) SEM images of C-PDA coated carbon fiber reinforced high-temperature composite. Adapted from Liu Y, Su C, Zu Y, Chen X, Sha J, Dai J. Ultrafast deposition of polydopamine for high-performance fiber-reinforced high-temperature ceramic composites. *Sci Rep*. 2022;12(1):20489. Creative Commons.[38] (**j**) SEM images of the PU-PDA. Adapted from Wang P, Zhang Y-L, K-L F, et al. Zinc-coordinated polydopamine surface with a nanostructure and superhydrophilicity for antibiofouling and antibacterial applications. 10.1039/D2MA00482H. *Mater Adv*. 2022;3 (13):5476–5487. Creative Commons.[39]

at treating bone diseases. However, these systems encounter limitations such as low loading capacity, limited physiological compatibility, degradability, and the propensity for rapid drug release by the systems. In recent years, numerous investigations have incorporated PDA as a composite component in biomaterials/drug delivery vehicles to augment drug loading capacities, bolster the trapping efficiency of cells in vivo, and address pathogen resistance. PDA-modified biomaterials have developed into various forms of applications, including nanotubes, nanospheres, and nanofilms, etc. (Table 1). Within the realm of orthopedic disease management, PDA-modified biomaterials are gradually becoming a major focus of research and application due to their multifunctionality and biocompatibility. The involvement of PDA can significantly enhance the anti-inflammatory, antibacterial, and antioxidant capabilities of orthopedic treatment strategies, and they notably improve the effectiveness of these strategies in promoting osteogenic differentiation and enhancing the activity of bone cells (Figure 4). The research and application of PDA span a diverse array of conditions, including bone injury repair,[40] bone regeneration,[41] osteoporosis,[42] and osteoarthritis.[43] Irrespective of the type of orthopedic condition, the onset age for bone diseases is increasingly younger,[44–46] with a concurrently diversifying patient demographic. The development of innovative drug systems utilizing PDA is poised to significantly enhance personalized treatment approaches for orthopedic diseases due to their high loading capacity and controlled release mechanisms. In the application of PDA coatings on implants for bone injury repair, the self-healing property is crucial. PDA holds significant promise in the field of self-healing materials due to its exceptional adhesion and intrinsic chemical structure, which enables the automatic formation of new cross-links. PDA can be combined with polymer matrices to create self-healing polymer materials used in various applications, such as electronic equipment and automotive parts. For instance, PDA particles and hindered urea bonds can be utilized as functional nanofillers and dynamic motifs, respectively, to produce dynamic cross-linked polyurea/PDA(DCPU/PDA) nanocomposites via facile in situ

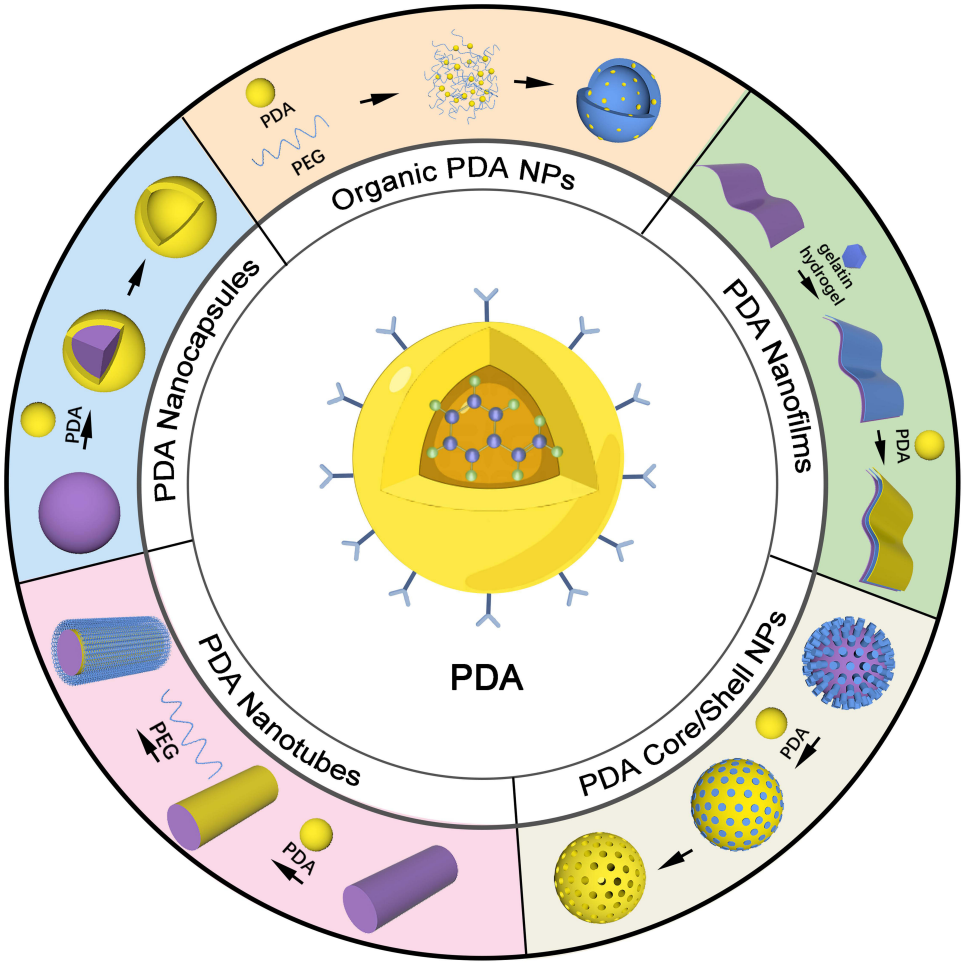

Figure 3 Classification of Polydopamine Nanoparticles. The diagram categorizes the various forms of polydopamine nanoparticles, including: organic PDA nanoparticles, PDA nanocapsules, PDA nanotubes, PDA core/shell nanoparticles and PDA nanofilms.

photoinitiated copolymerization.[47] These nanocomposites exhibit higher toughness and stretchability compared to pure DCPU. They self-heal rapidly and effectively in response to near-infrared light. Traditional thermo-responsive self-healing elastomers face challenges, including long repair times and limited self-healing sites. To address these issues, fast NIR light photo-responsive self-healing nanocomposites were fabricated by blending PDA particles into cross-linked polyurea containing sextuple H-bonds (SHBs) and hindered urea bonds (HUBs).[48] These composites show enhanced

Table 1 Advantages of Different Polydopamine-Based Applications for Orthopedic Diseases

Types	Advantages
PDA nanosphere shell	Characteristics of High-Loading, Sustained-Release Drug Carriers
PDA nanocapsules	The thickness of the material is controllable, enabling regulated drug release. Upon conjugation with branched chains, the system acquires enhanced bone-targeting capabilities, alongside notable antibacterial and antioxidant properties
PDA carbon/titanium nanotubes/nanorods	Regulates cell behavior to enhance osteogenic activity, thereby promoting the viability, adhesion, migration, and osteogenic differentiation of mesenchymal stem cells (MSCs) in vitro
PDA nanohydrogel	Enhances the mechanical properties, antibacterial activity, tissue adhesion, and osteogenic differentiation capabilities of hydrogels
PDA nanofilm coating	The microenvironment enhances MSCs' adhesion, proliferation, and osteogenesis, while concurrently downregulating pro-inflammatory cytokines in chondrocytes

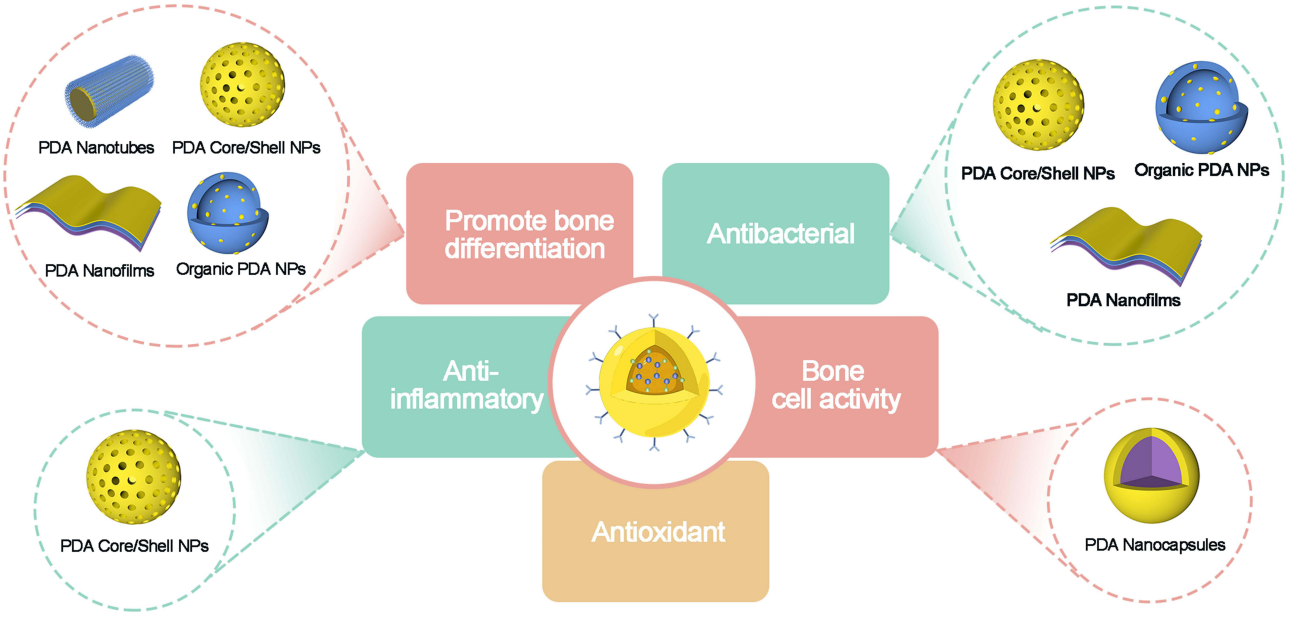

Figure 4 Application Spectrum of Polydopamine (PDA) in Orthopedic Disease Management: Material-Specific Utilizations and Therapeutic Directions.

mechanical properties and rapid photoresponsive self-healing capabilities due to the extensive H-bonds between PDA and polyurea fragments. In orthopedic applications, self-healing coating can alleviate corrosion, excessive degradation, and accidental scratching of metal-based implants. A previous study demonstrated the construction of a self-healing polymeric coating on Mg alloy, incorporating a stimuli-responsive drug delivery nanoplatform via spin-spray layer-by-layer (SSLbL) assembly. This nanocontainer system, based on simvastatin (SIM)-encapsulated hollow mesoporous silica nanoparticles (S@HMSs) modified with PDA and polycaprolactone diacrylate (PCL-DA) bilayer,[49] exhibited dynamic reversible reactions, fast and stable self-healing capacity, and excellent antibacterial properties under NIR irradiation. Additionally, it promoted cytocompatibility, osteogenesis, and angiogenesis. Collectively, PDA-based materials exhibit significant potential as self-healing materials and can play crucial roles across various fields. With technological advancements and increasing application demands, the prospects for PDA-based self-healing materials will continue to expand and deepen.

Polydopamine Core/Shell Drug Delivery Systems for Bone Disease Treatment

PDA can encapsulate a diverse range of drugs, enabling therapeutic efficacy at lower doses and minimizing side effects. The PDA spherical shell exhibits a porous structure, extensive specific surface area, and compact volume. Its interconnected pore architecture not only mimics bone composition but also aligns with the microscopic size and shape requisites for bone repair. Recent study demonstrated the Chitosan/Polydopamine/Octacalcium Phosphate (CS/PDA/OCP) microcarrier, which combines porous chitosan for cell adhesion and proliferation with octacalcium phosphate to mimic natural bone components.[24] Enhanced with PDA for superior adhesion, this microcarrier facilitates the formation of microstructures conducive to bone repair, establishing a three-dimensional microenvironment aimed at precisely repairing irregular bone defects. The PDA shell surface offers numerous active sites for additional modification,[50] This affects the mechanical properties, stability, and even optical properties of the material. In PDA-NPs, π-π interactions contribute to the formation of hierarchical structures and affect the ability of the material to adhere to various surfaces.[51] Thus this π interaction anchors the peptide or protein to the nanoparticle.[52] Zhang et al encapsulates the NO precursor N, N'-disec-butyl-N,N'- dinitroso-1, 4- phenylenediamine (BNN6) within a β-CD hydrophobic cavity on PDA-coated BG NPs.[53] This setup is designed to enable NIR-triggered NO release, promoting antibacterial effects and osteogenic differentiation of MSCs. PDA-coated inorganic nanoparticles, including gold nanoparticles, magnetic iron oxide, and non-metallic nanoparticles, can be tailored for sustained and controlled release by incorporating active ingredients. PDA itself is not a therapeutic agent but must be conjugated with drugs, photosensitizers, metals, or metallic nanoparticles to

enhance their functionality. The combination of PDA with metallic nanoparticles, such as gold, platinum, silver, iron oxide, and zinc oxide, synergizes cancer treatment and imaging of cancer cells, which are valued for their magnetic properties, chemical stability, tunable morphology, and ease of surface functionalization.[54] The functional groups on PDA's surface (catechol, carboxylic, amine, and imine) enable the binding of specific molecules or the loading of transition metal ions. For example, iron oxide nanoparticles can serve as contrast agents in magnetic resonance imaging (MRI), while zinc oxide enhances cancer cell killing, and gold nanoparticles improve cancer cell death.[54–57] Numerous in vitro studies have demonstrated that iron oxide nanoparticles promote osteoblast differentiation and inhibit osteoclast formation, while in vivo studies show that they accelerate bone defect repair and prevent bone loss.[55] For instance, previous studies reported that iron oxide nanoparticles coated with a PDA film acquire superior biocompatibility and multifunctionality.[56] A magnetic iron oxide/polydopamine coating significantly improves osteogenesis in 3D-printed porous titanium scaffolds under a static magnetic field.[57] Moreover, PDA combined with iron oxide nanoparticles can provide precise targeting to specific areas under the influence of a magnetic field. Researchers used Fe_3O_4@PDA to label MSCs, guiding them to pain-related response sites in spinal cord segments under magnetic control, thereby enhancing MSC homing and gathering for effective repair.[58] Most applications of PDA-coated iron-oxide NPs focus on cancer treatment, also for bone tumor diseases. For instance, to improve the efficacy of cisplatin in treating cisplatin-insensitive osteosarcoma, a study reported using iron-polydopamine coated multifunctional nanoparticles (SiO2@PDA/Fe^{3+}-FA).[59] These PDA-modified NPs have demonstrated high drug delivery efficiency, precise pH-responsive drug release, good biocompatibility, effective tumor targeting, and satisfactory photothermal efficiency, making them an effective tool for synergistic therapy in combination with NIR irradiation for drug-resistant tumors.

On the other hand, near-infrared (NIR) light-triggered shape memory materials show significant potential in biomedical applications compared to traditional heat-triggered shape memory polymers. For instance, a study reported the synthesis of polycaprolactone (PCL)–PDA polyurethanes using PCL, PDA nanoparticles (PDA NPs), hexamethylene diisocyanate (HDI), and 1,4-butanediol (BDO). These polyurethanes exhibited rapid shape recovery under NIR light due to the photothermal effect of PDA. The PCL–PDA polyurethanes demonstrated excellent in vivo NIR light-triggered shape memory performance under an 808 nm laser with low intensity, highlighting their potential for biomedical implant applications.[60] Another study reported the preparation of HNTs@PDA through in-situ free radical polymerization of acrylamide in a mixture of Laponite-RD, HNTs@PDA, and gelatin. HNTs@PDA, synthesized via oxidative polymerization of DA on the surfaces of HNTs, served as a superior photothermal agent for light-responsive hydrogels. The NIR-triggered shape recovery process of the HNTs@PDA hydrogel was notably rapid. These reinforced hydrogels, with superior mechanical properties, NIR-triggered shape memory, and self-healing ability, exhibit promising applications in biomedical materials.[61] Smart self-healing coatings have also garnered significant interest due to their switchable and desirable functionalities in response to external environmental changes. An eco-friendly smart self-healing coating with NIR and pH dual-responsive superhydrophobic and anti-biofouling properties was fabricated by mixing biomimetic stimuli-responsive mesoporous PDA microspheres (polydimethylsiloxane-loaded mesoporous polydopamine microspheres, abbreviated as P-PDMS@MPDA MSPs) with waterborne resin and hydrophobic nanoparticles.[62] This coating demonstrated self-healing of its superhydrophobicity and active anti-biofouling properties under NIR or pH stimuli due to the release of low-surface-energy PDMS from the P-PDMS@MPDA MSPs, showing excellent self-healing and biological properties. By adjusting the NIR light intensity, PDA microspheres can achieve precise temperature control of shape memory materials, enabling the material to undergo shape changes within the desired temperature range.[63,64] Not only the toughness and strength of PDA particle-filled polyurethane composites (SMPU-PDAPs) have been significantly improved with the addition of PDAPs.[65] The tensile stress of damaged nanocomposites can also be recovered using the heat energy generated by near-infrared lasers.[66] This capability holds significant promise for biomedical applications necessitating precise manipulation, such as minimally invasive surgical instruments and implantable medical devices. For instance, strontium ions, which promote osteogenesis and angiogenesis, are stably incorporated into PDA microspheres due to PDA's coordination reaction,[40] optimizing the osteogenic potential of strontium ions while mitigating the adverse effects of high concentrations. Xiong Wei et al introduced a composite microsphere that can release both the antibacterial agent berberine (BBR) and the bone-strengthening drug naringin (NG), utilizing the simple synthesis of PDA coatings.[41] This microsphere demonstrates excellent biocompatibility and degradability, making it

a promising option for treating bone defects at infected sites. Additionally, Leveraging the photothermal conversion capabilities of PDA particles and the concurrent release of adjuvants from mesoporous silica shells, researchers have developed PDA (core)-mesoporous silica (shell) nanocapsules aimed at tumor photothermal therapy.[67] PDA serves as a biodegradable photothermal agent with excellent biocompatibility, playing a key role in anti-tumor activity. PDA is renowned for its exceptional near-infrared (NIR) absorption properties, which are crucial for biomedical applications involving irradiation-sensitive materials. Here are the fundamental reasons for its effectiveness. PDA possesses a high concentration of aromatic rings, creating an extensive π-π conjugated system. This conjugation enables the material to absorb light energy across a broad wavelength range, including the NIR region.[11] The π-π system facilitates electron delocalization within the molecule, resulting in more efficient light absorption and energy transfer. PDA is abundant in radicals and semiquinone structures, which exhibit strong NIR absorption. These structures provide additional energy levels and electron transition pathways, significantly enhancing PDA's NIR absorption capabilities. The high degree of polymerization and tight molecular packing in PDA amplify intermolecular interactions. This packing not only supports efficient light absorption and conduction, but also enhances the material's stability and thermodynamic properties, thereby improving NIR absorption efficiency. The electronic structure of PDA can be optimized by adjusting synthesis conditions such as pH, temperature, and reaction time. For instance, doping PDA with metal ions or other functional groups can modify its electronic energy levels and optical properties, optimizing NIR absorption.[68] Conjugated π-electronic systems in PDA molecule-coated gold nanoparticles exemplify how such modifications can effectively enhance NIR light absorption.

PDA is highly multifunctional and tunable. Chemical modifications can introduce various functional groups, altering its optical properties. This tunability ensures that PDA performs exceptionally well in diverse NIR applications. As a biomimetic material, PDA offers excellent biocompatibility and environmental friendliness. Its safety and effectiveness in NIR absorption make PDA an ideal choice for biomedical applications involving irradiation-sensitive materials.

PDA Nanocapsules

PDA-based nanocapsules represent a prevalent form of drug delivery system. Previous report demonstrated PDA nanocapsules exhibit a high loading capacity, leveraging curcumin (CCM) and monoconcosaccharide interactions to create supramolecular structures. Upon modification with polydopamine and incorporation of drugs similar in polarity to CCM, the capsules maintain integrity while facilitating the formation of dual-drug nanoparticles for bidirectional drug delivery.[69] The capsule's drug release rate can be precisely adjusted through the manipulation of PDA thickness, enabling controlled drug delivery. This feature is particularly useful for administering anti-inflammatory agents like curcumin in the treatment of osteoarthritis.[70] Furthermore, the synthesis of PDA nanocapsules were through ammonia- induced PDA polymerization for coating silica mesoporous nanoparticles, as well as the formation of PDA nanocapsules by template removal following water dispersion,[71] have been effectively employed as nanoparticles in drug delivery. Several studies have coated $Gd_2(CO_3)_3$ cores with PDA to create nanoparticles, to which a cartilage-targeting peptide was modified, and hesperetin was incorporated to establish a cartilage-specific functional drug delivery system. Experimental results have shown high affinity of this system for cartilage and its potential applications in magnetic resonance imaging (MRI).[72] Research has shown that folic acid (FA) is an effective targeting agent for arthritis. By conjugating FA to PDA, a targeted nanodrug delivery system can be developed to target the arthritis site. Utilizing the pH-responsive mechanism, the NH_2-PEG-FA ligand is synthesized to actively target OA, with size optimization ensuring full utilization of the EPR effect instigated by inflammation, thereby facilitating both effective passive accumulation and active targeting in OA treatment.[73] Yun Wang et al developed a mesopore-based microsphere encapsulating a drug clearance delay system (RCGD423@MPDA), employing MPDA) loaded with the small molecule RCGD423 for modulating the inflammatory response. This system demonstrates superior drug loading capacity, exhibiting longer drug retention and more stable, prolonged drug release compared to traditional poly (lactate-co-glycolic acid).[74] In a rat model of osteoarthritis, administration of this system proved more effective in inhibiting cartilage damage and proteoglycan loss compared to treatments without the drug delivery system, suggesting its potential in mitigating the degenerative changes associated with arthritic cartilage. Mesoporous polydopamine nanoparticles are capable of extending the drug release cycle, thereby diminishing the need for frequent intra-articular injections. This presents significant benefits compared to the regular

administration of short-acting hyaluronic acid. Moreover, the ability to control drug release minimizes adverse drug reactions and mitigates the potential for drug resistance.

PDA Nanotubes

Recently, nanotubes formed by PDA coating (PDA@CNTs) have garnered considerable interest. Studies have demonstrated that PDA coating on carbon nanotubes significantly enhances cytocompatibility.[75] These PDA-coated carbon nanotubes have shown promise in various applications such as drug delivery, antimicrobial and antioxidant agents, and biological drugs.[76] Their porous and adjustable interfaces make PDA@CNTs a promising biocomposite with extensive biological, diagnostic, and therapeutic potential. For example, the integration of osteogenic growth peptide (OGP) with TiO_2 nanotubes via PDA has been found to promote cell proliferation and differentiation.[77] Moreover, PDA-coated TiO_2 nanotubes-MoS_2/PDA-LL-37 demonstrate potent antimicrobial activity and enhance bone regeneration when exposed to near-infrared irradiation.[78] PDA-modified titanium (Ti) substrates enhance the viability, adhesion, migration, and osteogenic differentiation of MSCs.[79] Additionally, biofunctionalized TiO_2/MoS_2/PDA nanotube coatings on Ti implants have been developed to stimulate osteogenic activity, notably through the simulation of MC3T3-E1 osteoblasts' differentiation and the upregulation of Runx2.[80] Recent clinical studies indicate that the majority of Ti implant failures are due to bacterial adhesion. Introducing a PDA bioactive coating on nanotubes can significantly mitigate implant infection issues associated with Ti implantation, due to the inherent antibacterial properties of PDA. Furthermore, PDA is capable of immobilizing growth factor proteins, for example vascular endothelial growth factor (VEGF),[81] a property utilized in the PDA coating of TiO/MoS/PDA nanotubes to augment osteoinductivity.[82] Strontium nanotubes/PDA arginine-glycine-aspartic acid coatings have also been employed on Ti_6Al_4V materials, significantly enhancing MSC adhesion and promoting MSC differentiation, thereby demonstrating outstanding bone-regeneration potential.[83] The recent proposition of composite artificial periosteum offers an innovative solution to the significant clinical challenge posed by periosteal defects.[84] Leveraging PDA's high tissue adhesion properties, the composite artificial periosteum features ends cross-linked with a PDA hydrogel layer of carbon nanotubes (CNTs), embodying a design that mimics the directional arrangement of periosteal collagen fibers. This construction ensures adequate mechanical strength and the desired directional nanotopological surface. Highlighting the potential of PDA nanotubes in orthopedic applications, their role in promoting bone regeneration demonstrates promising prospects.

Advancements in PDA Nanoparticles Crosslinked / Integrated with Other Materials

PDA nanoparticles can be cross-linked with acrylamide and additional polymers to fabricate hydrogels,[85] PDA nanoparticles crosslinked with acrylamide and other polymers leverage the high adhesion properties of polydopamine to enhance mechanical properties and biocompatibility. One notable example is the PAM/BA-Ag@PDA hydrogel, created by in situ polymerizing acrylamide (AM) with N,N′-bis(acryloyl)cystamine (BA), dynamically crosslinked with silver-modified polydopamine (PDA) nanoparticles.[86] This multi-functional hydrogel sensor exhibits significantly enhanced tensile and compressive strength, reduced hysteresis, improved conductivity, and excellent near-infrared (NIR) light-triggered self-healing abilities compared to traditional polyacrylamide (PAM) hydrogels. As a strain sensor, PAM/BA-Ag@PDA hydrogel demonstrates good sensitivity, rapid response time, and high stability. Previous studies have reported the fabrication of self-healing and adhesive, electrically conductive, and biocompatible PAM nanocomposite hydrogels. These are produced via in situ polymerization of acrylamide in the presence of polydopamine-modified carbon nanotubes (PDA@CNTs). Such hydrogels function as flexible strain or pressure sensors, effectively detecting human motions, identifying materials and their shapes, and transmitting health information,[87,88] This strategy was also been extensively explored in the development of conductive hydrogels for biosensors. Additionally, PDA's photothermal effect significantly improves antimicrobial applications. One study demonstrated that PDA's photothermal properties can induce heat production to cause bacterial death under near-infrared light radiation. This research involved the uniform coating of PDA and polypyrrole (PPy) onto poly(l-lactide) (PLLA) nanofibers via in situ polymerization, resulting in a novel PPy/PDA/PLLA three-layer core-shell structure. The homogeneously coated PPy and PDA layers significantly increased hydrophilicity, conductivity, and near-infrared photothermal antibacterial properties, while also providing antioxidant capacity and reactive oxygen species (ROS) scavenging ability.[89] Given their high biosafety, numerous covalent modification sites, and superior adhesion, hydrogels derived from cross-linked PDAs are ideally suited for biomedical applications. Hydroxyapatite

Table 2 Effects of PDA Combined with Other Materials in Preventing and Treating Orthopedic Diseases

PDA+Other Materials	Effect
PDA+NO Precursors	Enhance antibacterial, promote the osteogenic differentiation of mesenchymal stem cells (MSCs).[53]
PDA+Mesoporous silica (shell)	Sustained-release drug carriers.[67,71]
PDA+PEG-FA	Target osteoarthritis.[73]
PDA+TiO_2	Promote new bone formation.[35,77,78,80]
PDA+Ti_6Al_4V	Enhance the attachment and differentiation of MSCs, and promote bone reconstruction and regeneration.[83,104,105]
PDA+CNTs	Enhance mechanicality, fix the artificial periosteum, and promote bone regeneration.[75,88]
PDA+Hap	Increase biocompatibility and promote osteogenic differentiation,[26,90,99,100,106–108]
PDA+Ti	Enhance resistance to MRSA infection,[79,94]
PDA+HA	Enhance mechanical properties, cell affinity, tissue viscosity.[98–100]
PDA+PVDF+$BaTiO_3$	Promote pre-osteoblast viability.[109]
PDA+mPEG-NH_2	Down-regulation of cartilage pro-inflammatory factor.[110]
PDA+PCL	Promote stem cell osteogenic differentiation.[60,111–113]

(HAP), a critical component of human bone, significantly affects cell behavior, including adhesion, proliferation, and differentiation, and is commonly employed in tissue regeneration. However, HAP fragments may trigger inflammation, impeding osteoblast growth.[26] PDA-modified HAP nanoparticles can surmount the inherent limitations of HAP-based materials in tissue repair. Specifically, PDA-mediated incorporation of bioactive peptides or proteins (such as bone morphogenetic protein-2) onto HAP nanoparticles has been shown to enhance their biocompatibility and promote osteogenic differentiation.[90]

Numerous studies have utilized PDA NPs in combination with various active compounds to promote osteogenic differentiation,[91–95] and exhibit antimicrobial properties.[96,97] Additionally, PDA can produce diverse effects in the prevention and treatment of orthopedic diseases when combined with other materials. (Table 2). In clinical surgery, researchers have developed a multifunctional material, Ti-PDA@SNP-OGP, to address the antimicrobial deficiencies of titanium implants.[94] This composite integrates mesoporous polydopamine nanoparticles, the nitric oxide-releasing donor sodium nitroprusside (SNP), and osteogenic growth peptide (OGP) onto titanium implants. The goal is to effectively combat methicillin-resistant Staphylococcus aureus (MRSA) infections during implant replacement procedures. Furthermore, PDA acts as a functional modifier for cell-loaded hydrogels, enhancing the properties of the base materials. Pure hyaluronic acid (HA) hydrogels exhibit suboptimal mechanical properties. However, when HA hydrogels are modified by cross-linking with PDA, their mechanical properties are enhanced, and the critical gelation concentration is lowered.[98] This modification improves cellular affinity and tissue adhesion, while also imparting free radical scavenging and antibacterial capabilities to the hydrogel. Additionally, incorporating HAP into PDA-HA further boosts the hydrogels' osteogenic differentiation capabilities and exhibits significant antimicrobial potential.[99,100] Synthetic polymers, such as polylactic acid (PLLA), polyglycolic acid (PGA), and polycaprolactone (PCL), serve as versatile substitutes for natural bone. These polymers exhibit notable biocompatibility, flexibility, and stability within the human body. However, these materials face challenges, including biologically inert surfaces and insufficient mechanical strength. To address these issues, recent studies have focused on functionalizing synthetic polymers notably through employing PDA as a nanoparticle coating.[101,102] This approach leverages PDA's excellent adhesion and biocompatibility to enhance the surface activity of polymers and improve their mechanical strength, including tensile properties. Nanoparticles, integrated with PDA to create "building blocks" are rapidly advancing across various domains, including pharmaceuticals, inorganic compounds, proteins, and metal-organic frameworks (MOFs).[103]

Evaluating the Impact of PDA Nanofilm Coatings on the Bone Reconstruction and Regeneration Microenvironment

The significant adhesive properties of PDA allow it to be applied to a wide range of material surfaces. To date, utilizing PDA has been recognized as a straightforward and efficacious strategy for modulating interfaces in biological tissue engineering. This coating can directly transmit biological signals to cells,[114,115] enhance the hydrophilicity of the interface, and promote cell diffusion and attachment.[116,117] Moreover, a specific surface roughness of the coating augments protein absorption, consequently enhancing cell adhesion.[118] Furthermore, early studies have demonstrated that surface roughness at the micron and submicron levels significantly facilitates osteoblast differentiation.[119] PDA nanofilm coatings have been extensively studied in

the treatment of orthopedic diseases,[49,120–124] and for antimicrobial purposes,[125–128] with strategies being progressively refined. The surface hydrophilicity of bone repair materials is closely related to their biocompatibility and cell adhesion. PDA contains substantial numbers of hydrophilic groups, such as amino and hydroxyl groups, which can be bound to hydrophobic surfaces, thereby effectively improving the hydrophilic properties of bone repair materials.[129] For example, polycaprolactone (PCL) is a commonly investigated material in the field of bone tissue engineering; however, its application is limited by its bioactivity. Numerous studies have reported using PDA to coat PCL surfaces, either in powder form or on scaffolds. PDA-coated PCL powder used for scaffold fabrication provides more hydrophilic surfaces for cell adhesion and growth than pure PCL scaffolds due to the presence of amino and hydroxyl functional groups.[130] A study demonstrated the effects of PDA coating under different conditions, including titanium (Ti), PDA-coated Ti samples, and PDA-coated Ti samples either stored for up to two weeks at room temperature or heated at 121 °C for 24 hours. The results emphasized that PDA coating heated at 121 °C for 24 hours did not impair the water contact angle and increased cell proliferation for both hDFs, HaCaTs, and MC3T3-E1 cells compared to pristine PDA. This underscores the importance of post-treatment and shelf-time for PDA coatings.[131] Additionally, moderately hydrophilic materials can adsorb an optimal layer of proteins, fostering a conducive microenvironment for cell adhesion at the cell-material interface. Based on the rapid formation and accumulation of PDA nanoparticles, some researchers have used superhydrophilic PDA coatings synthesized in situ by Ag NPs in the presence of sodium periodate, which effectively reduced the non-specific adsorption of proteins.[132] By using PDA materials, can regulate cell behavior at the interface of biomaterials, which can be better used for wound healing, including bone trauma treatment.[133] A novel Mn_3O_4@PDA@Pd-SS31 nanozyme targeting mitochondria was designed to reverse mitochondrial dysfunction and inhibit inflammation.[134] PDA has the ability to bind to various serum proteins, resulting in substrates that enhance cell attachment. On this hydrophilic matrix, cells exhibit excellent attachment, expansion, proliferation, and differentiation properties.[135] This is especially important in tissue engineering, regenerative medicine, and biosensor applications. Polydopamine nanopreparations when coated with polyvinylidene fluoride (PVDF) and integrated into nanocomposites with BaTiO, have been found to significantly promote cell viability and pre-osteoblast migration in vitro, as well as accelerate the formation of biomineralized apatite layers.[109] The study demonstrated that mPVDF-BT coatings, leveraging the adhesion and biocompatibility of PDA, were applied to rough Ti_6Al_4V biomaterial surfaces. PDA addresses the bio-inertness and enhances the long-term stability of Ti_6Al_4V, with the coated biomaterial showing potential in promoting bone reconstruction and regeneration. Ko et al developed a functional electrospun silk fibroin (SF) nanofiber scaffold that underwent conversion into two-stage HAP particles through PDA coating. Subsequent studies validated that HAP coated with PDA facilitates osteogenic differentiation and boosts bone formation both in vivo and in vitro.[106,107] Furthermore, a coating on Hap/polyamide-66 (HA/P66) substrates was devised through a PDA-assisted biomimetic process, enhancing HAP dispersion and accelerating osseointegration.[108] The surface roughness and bioactive properties of the HAP coating are likely contributors to the osteogenic differentiation supported by Hap-PDA-HA/P66 substrates in mouse MSCs. Numerous studies have highlighted the ability of PDA to enhance cell adhesion and osteocyte differentiation.[136–138] The primary mechanism underlying osteogenesis promotion by PDA is attributed to its enhancement of the material's hydrophilicity and surface roughness, thereby improving cellular access to the biomaterial surface. Moreover, PDA facilitates the integration of bioactive factors, including bone morphogenesis protein-2 (BMP-2) and vascular endothelial growth factor (VEGF), into its multilayer coatings.[139] This approach enhances osteoconductivity for cellular protein binding, further encouraging osteoblast adhesion, proliferation, and differentiation.[140] Research has revealed that PDA nanocoatings on biomaterials not only facilitate mesenchymal stem cell attachment but also trigger human cell reprogramming and sustain the long-term self-renewal of human pluripotent stem cells (hPSCs).[141] Investigations have demonstrated that methoxypolyethyleneamine (mPEG-NH_2)-modified PDA NPs effectively downregulate pro-inflammatory cytokines in chondrocytes, thereby mitigating cartilage and subchondral bone inflammation in rat OA models.[110] Moreover, the PDA NPs coating has been optimized for application on electrospun PCL fiber membranes, creating an optimal microenvironment for directing local stem cells towards osteoblast differentiation. Additionally, this fiber membrane acquires degradability from PDA NPs,[111] enhancing hydrophilicity and cytocompatibility alongside its inherent biodegradability. The findings indicate that PDA/PCL fiber membranes facilitate the diffusion, proliferation, and osteogenic activity of human mesenchymal stem cells (hMSCs) in vitro, exhibiting dose-dependent effectiveness. Significantly, in a mouse model presenting a critical-sized skull defect, the osteogenic differentiation of hMSCs was notably enhanced by PDA-modified biomaterials. PDA-based materials have demonstrated the capability to

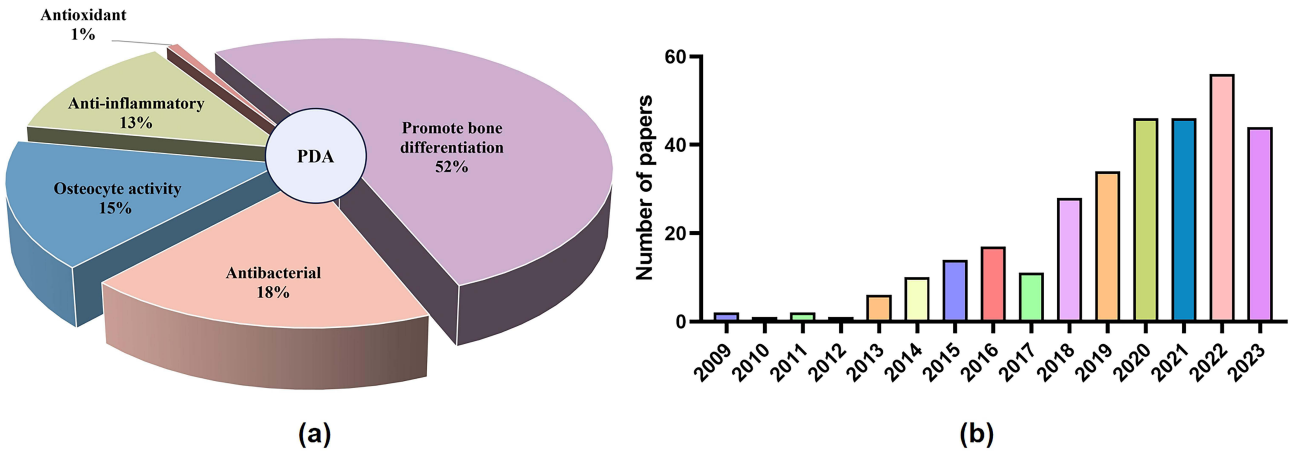

Figure 5 Distribution of PDA Applications and Research Trends. (**a**) Proportional representation of PDA application directions. (**b**) Annual distribution of orthopedic-related publications of PDA, including a total of 4 articles in 2024 currently.

eliminate reactive oxygen species (ROS) both in vitro and in vivo,[5] with prior research delving into the mechanism of PDA NPs as ROS scavengers in dental applications, yielding promising outcomes. In vitro experiments revealed that PDA NPs act as scavengers for hydroxyl (HO) and superoxide radicals, achieving a scavenging rate of up to 90% for HO at a concentration of 0.1 mg/mL, with superoxide· being entirely eliminated from the system as anticipated. Fluorescence imaging and antioxidant therapy experiments for in vivo ROS clearance showed that PDA NPs, at a dosage of 0.2 mg/site through subgingival injection, could efficiently eradicate local ROS and significantly mitigate periodontal inflammation. Furthermore, spectroscopy and additional in vitro experimental findings offer robust evidence that PDA NPs can eliminate various ROS and suppress ROS-induced inflammatory reactions,[142] suggesting that PDA-NPs create an anti-inflammatory milieu conducive to bone reconstruction and regeneration.

Summary and Outlook

Bone, a critical tissue and organ for normal motor function in humans, has made the treatment of bone diseases a significant focus in medical field. Currently, biomaterials for bone disease treatment face challenges such as instability of drug-loaded materials, insufficient cell adhesion, reduced biocompatibility, and potential human toxicity from degradation products. Extensive research has demonstrated that biomaterials those modified with PDA surface engineering, possess antibacterial, anti-inflammatory, and antioxidant properties, can induce osteogenesis, enhance hMSC bone differentiation, promote osseointegration, and accelerate new bone formation. Since 2009, polydopamine nanoparticles have gained popularity among orthopedic treatment researches, with 52% of studies concentrating on osteogenic differentiation. This is followed by 18% on antimicrobial effects, 15% on enhancing cell activity, 13% on anti-inflammatory effects, and the rest on antioxidant properties (Figure 5). Research publications involved PDA on bone diseases treatment have significantly increased post-2017, peaking in 2022 with the highest number in recent years, predominantly focusing on promoting osteogenic differentiation (Table 3 and Figure 5). Polydopamine nanopreparations are becoming one of the key biomaterials for the treatment of bone diseases. As a drug delivery system, nanopreparations

Table 3 Summary of Research Publication on the Application of Polydopamine (PDA)

PDA Applications	Total Publications
Enhances Osteogenic Differentiation[5,24,26,40–42,49,57,72,77,78,80,82–84,90–95,99,100,104–106,108,112,119–124,126,129,138,140,143–292]	188
Enhances Antimicrobial Activity[53,76,80,81,90,96,97,99,100,112,113,125–128,211,223,278,293–340]	66
Stimulates Relevant Cellular Functions[24,79,106,136–141,146,160,169,183,188,189,193,210,215,221,222,226,227,240,264,265,285,287,341–370]	57
Enhances Anti-inflammatory Properties[43,70,72,73,94,97,110,146,228,249,269,371–406]	47
Enhances Antioxidant Activity[76,407,408]	3

incorporating PDA exhibit high drug loading capacity, effective sustained-release properties, and delayed drug clearance, offering significant potential to address current challenges in the clinical treatment of bone diseases. Furthermore, PDA can functionalize a diverse array of nanomaterials, facilitating the development of a PDA-based multifunctional platform for targeted or synergistic therapy. Despite the limited clinical translation of many biomaterials in the realm of bone disease treatment, PDA preparations, whether employed as biologically functional additives or drug carriers, are expected to play a more significant role in managing and preventing orthopedic disorders.

Data Sharing Statement

The data presented in this study are available in this article.

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Disclosure

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