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Mussel-inspired biomimetic adhesive coatings for food preservation: A review

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Abstract

In recent years, taking inspiration from mussel-an underwater expert, various packaging alternatives including adhesive coatings are being developed for food preservation to ensure food availability for everyone. The extraordinary adhesion, exhibited by mussel, is mainly offered by mussel foot protein containing catechol groups. This catechol-based chemistry not only improves adhesion, but also helps in imparting antimicrobial, antioxidant, and UV blocking properties to packaging materials for increasing the shelf-life of food items. Herein, we first present an overview of catechol-based chemistry followed by a discussion involving a combination of catechol and its derivatives with various biodegradable polymers and nanomaterials. Further, we summarize recent efforts of developing mussel-inspired catechol-based coatings for food preservation, keeping minimum environmental impact in mind. Finally, we discuss various challenges and opportunities existing in this area for successful commercial utilization of such biomimetic coatings in the future.

Keywords: Mussel, Biomimetic, Coating, Packaging, Shelf-life

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1. Introduction

World population is estimated to touch ≈9.8 billion by 2050 making food security a prime concern.[1] According to a United Nations' report ≈1 billion tons food was wasted in 2022.[2] Further, it has been reported that approximately one third of the fresh vegetables and fruits get spoiled before being eaten by the consumer.[3] This situation of food wastage is very critical and it affects the economy of every country, thus requiring technological interventions to ensure food availability for everyone.[4]

In quest of achieving the global sustainable development goal of "Zero Hunger" put forward by United Nations, food packaging plays a crucial role.[5] Packaging acts as a physical barrier safeguarding the food from outside contamination.[6] However, most of these packaging materials are manufactured using petroleum-based polymers leading to negative impact on the environment caused by landfill and incineration.[7] It has been reported that ≈141 million tons of plastic waste is generated every year by packaging sector.[8] This troublesome situation has sparked a surge in recent years to explore packaging materials that are biodegradable and can address different aspects of food packaging.[9] Such packaging alternatives including active packaging, intelligent packaging, and edible coatings can help in protecting the food from microbial spoilage or oxidation that can occur during storage, transportation, and purchase, while sensing the loss of freshness over time.[10-17]

Development of packaging materials require a polymeric matrix which is either made up of a single polymer or prepared by blending different polymers together. In this regard, natural polymers (e.g., polysaccharides, proteins) have emerged as an excellent choice of matrix owing to their inherent biocompatibility and biodegradability.[18] Further, polymer matrix is often loaded with active components such as nanomaterials and essential oils to impart active properties to the packaging.[19,20] Here, homogeneous mixing of different polymers and

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nanomaterials is very crucial to achieve the desired physicochemical properties. Additionally, in the case of edible coatings, satisfactory adhesion of the coating solution on the surface of fruits and vegetables is critical to avoid dripping so that uniform coverage can be achieved.[21]

Naturally existing mussel is known to be an "underwater expert" owing to its very strong adhesion on a variety of surfaces facing shear stress caused by flowing water.[22] This extraordinary adhesion is achieved by mussel foot protein containing dopamine having functional groups like catechol, amine and imine. These functional groups have been reported to improve adhesion and facilitate metal chelation while imparting antimicrobial, antioxidant, and UV blocking properties to materials. Based on the above-mentioned remarkable properties, mussel-inspired catechol-based chemistry can act as a promising platform in packaging sector for improving the shelf life of food items.[23-26]

In the last decade, a plethora of articles dealing with different aspects of food packaging materials have been reported in the literature.[27-32] Also, there has been a significant surge in efforts on exploring mussel-inspired catechol-based chemistry for biomedical applications.[33] However, the utilization of this mussel-inspired chemistry in packaging sector is still in its nascent stage and a detailed review on how it's potential can be harnessed for improving the shelf-life of food products is missing. Hence, herein, we first provide the readers a brief overview of catechol-based chemistry and its derivatives. Next, we discuss various approaches for utilizing catechol-based chemistry in combination with various polymers and nanomaterials. Further, we discuss recent advancement in utilization of mussel-inspired catechol-based coatings for food preservation while simultaneously minimizing the adverse impact on the environment. Finally, we summarize the challenges existing in this area that must be addressed to ensure the successful widespread application of such coatings in the future.

2. Mussel-inspired chemistry

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Mussel byssus releases mussel foot protein which is rich in 3,4-dihydroxy-l-phenylalanine (DOPA).[34] The secret of mussel's wet adhesion lies in the oxidation of catechol groups in basic medium followed by crosslinking with the amine groups of the secreted proteins. Taking inspiration from mussel foot protein, in 2007, Lee *et al.* historically utilized dopamine (DA) as a coating material which was practically applicable to all the tested substrates, rendering it as the most celebrated wet adhesion material in various areas of science.[35]

2.1. Catecholamines and catechol derivatives

Based on the site of chemical moiety incorporation in DA and the type of chemical moiety, different chemical analogues of DA can be made which have unique physicochemical attributes **(Scheme 1)**. DOPA, the most common derivative, acts as a precursor of DA in humans **(Scheme 1A, B)**. Presence of –COOH group at R1 position is the characteristic feature of DOPA and it is naturally synthesized by catalytic action of tyrosine hydroxylase enzyme. Different analogues of DOPA such as L-DOPA, CysteinylDOPA help in the formation of different pigments in living organisms **(Scheme 1B)**. In humans, L-DOPA is used in eumelanin formation via dihydroxyindole (DHI), which is responsible for the black color of skin and hair.[36] CysteinylDOPA is used for the formation of pheomelanin which provides red color to lips.[36] A combination of both L-DOPA and cysteinylDOPA is responsible for neuromelanin formation in brain.[36] Coating of polyDOPA on substrates has been reported to be less favorable as compared to polydopamine (PDA) owing to the electrostatic repulsive forces existing between the nearby –COOH groups.[37] To address this, high ionic strength conditions have been used to successfully coat polyDOPA on metals and polymeric substrates.[38] Addition of –OH group at R2 position results in another DA derivative known as norepinephrine (NE) which naturally exists as a neurotransmitter **(Scheme 1C)**. Oxidative polymerization of NE leads to very uniform, smooth, and biocompatible coatings. It has been reported that under similar

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coating conditions, polymerization rate of NE is slower than PDA. Additionally, the thickness of NE coating (≈20 nm) is lesser than PDA coating (≈40 nm).[39] NE based coatings have been successfully developed on a variety of substrates ranging from glass to polymers.[40]

Derivatization of $–NH_2$ group (R3 site) is one of the most often used methods for incorporating desired features into the coating. This derivatization hampers indole formation via cyclization, however, conjugation of catechol moieties facilitates the coating of substrate. Various substituents like 2-bromoisobutyryl, methyl, pyrrole, pyridine, methacrylate have been utilized for developing functionalized coatings **(Scheme 1D)**.[40, 41]

Incorporation of various substituents such as $–NO_2$, –Cl, –Br, –OH in the benzene ring (R4 site) also generates a range of DA analogues with unique properties. These derivatives including 6-nitrodopamine, 6-nitronorepinephrine, 6-chlorodopamine, 6-bromodopamine, 5-aminoethylpyrogallol, 5-hydroxydopamine can be utilized for preparing functional coatings with varying features **(Scheme 1E)**.[36, 42, 43, 44]. For example, 6-nitrodopamine is more oxidation resistant as compared to DA providing stronger adhesion on the surface of magnetic nanoparticles.[45]

Further, 5,6-dihydroxyindoles with hexamethylenediamine, 5,6-dihydroxy-3-ethinylindole with Ni^{2+}/O_2 or H_2O_2, and 5,6-dihydroxy-1H-indazole in basic medium/air have been utilized for coating the substrate **(Scheme 1F)**.[46] Apart from that, various plant-based polyphenols such as tannic acid (TA), gallic acid, and caffeic acid have also been widely explored as a coating material owing to the presence of catechol groups.[47-49]

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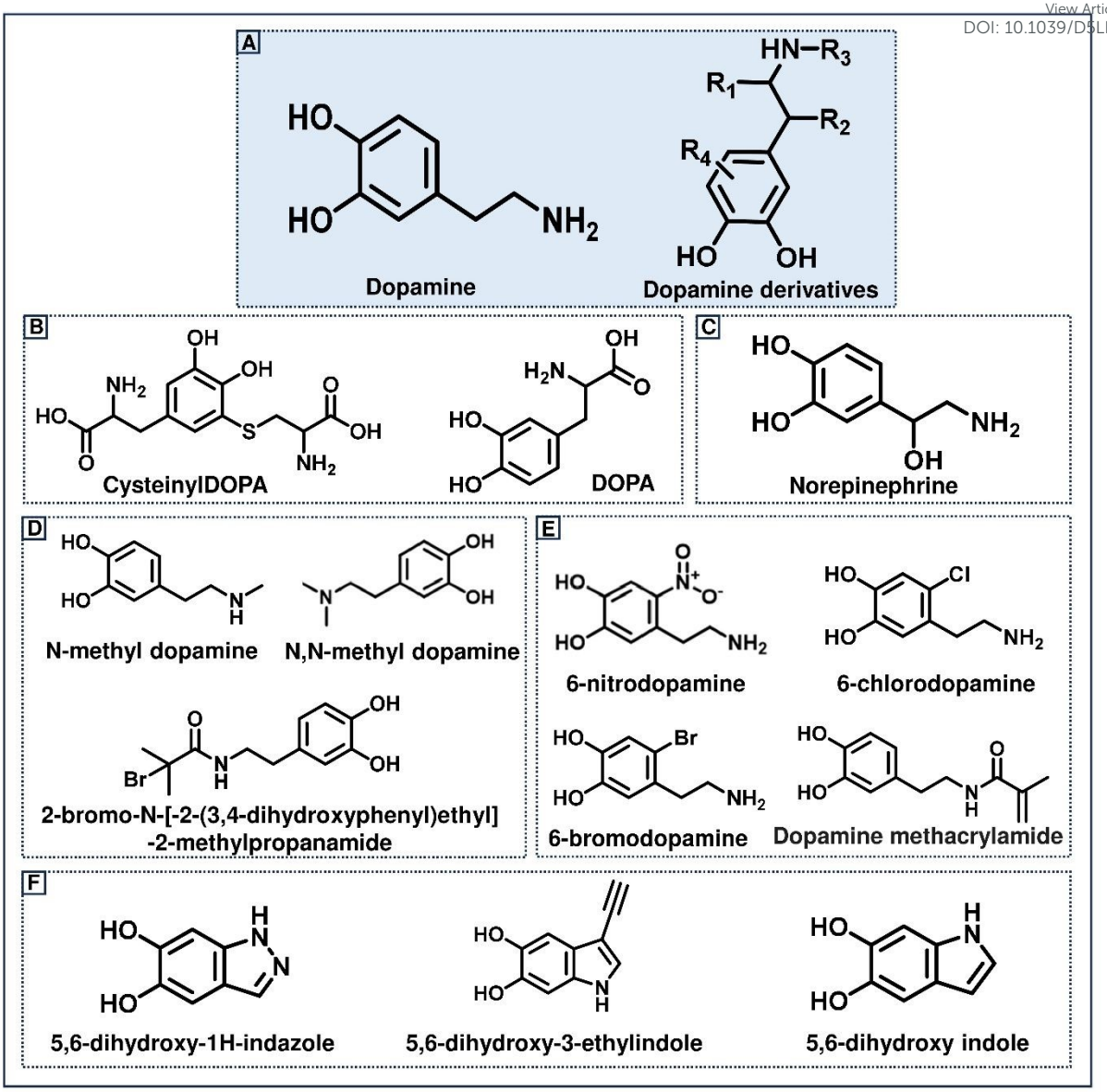

Scheme 1. (A) Chemical structures of dopamine and its derivatives prepared by chemical modifications at different sites depicted by R1, R2, R3, and R4. Functionalization at (B) R1 and (C) R2 positions of alkyl chain, (D) R3 position having amine group, and (E) R4 position having aromatic sites. (F) Dopamine derived heterocyclic compounds.

2.2. Catechol-mediated interfacial interactions in mussel adhesion

Adhesion is fundamentally determined by the intermolecular interactions that link atoms or molecules.[50] Traditional adhesives mostly function well on dry surfaces and have diminished efficacy in wet or underwater conditions. The loss of adhesion results from the development of

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a hydration layer that interferes with molecular interactions at the adhesive/adherent interface, ultimately causing bond failure.[50] Nature has produced various exemplary role models that inspire the design of efficient materials.[51] The marine mussel exemplifies an organism that has evolved the capacity to adhere firmly to submerged rock surfaces, enabling it to endure the intense forces of waves and predation.[52] The mussel achieves this remarkable function via disc-shaped, cell-free threads termed byssi, which effectively secure it to its designated substrate.[52] Each byssus generally consists of tens to hundreds of threads, with diameters ranging from 100 to 250 μm and lengths between 2 and 6 cm.[53] The distal end of each byssal thread is encased in a thin yet highly extensible protective cuticle, characterized by spontaneous stiffness and resilience.[54] The stem root at the foot's base functions to anchor the entire byssus. Mussels can rapidly discard their byssus as needed and regenerate a new one within hours.[55] Mussel adhesives exhibit superior strength and durability compared to the majority of synthetic adhesives. A study indicated that the force necessary to detach California mussels from the substrates ranged from 250 to 300 N, with an average separation force of 5 to 6 N per byssal thread.[56]

The enduring adhesive properties of mussels have led to extensive research aimed at outlining the mechanisms and chemicals involved in this natural phenomenon. The molecular mechanisms governing mussel adhesion in aquatic environments remain inadequately explained; however, evidence indicates that the secretion of specific proteins is crucial for mediating adhesive functions.[52] The mussel byssus consists of a complex arrangement of over 20 distinct protein threads, with six (mfp-1 to mfp-6) identified as major mussel foot proteins (MFPs) that have been extensively researched for their significant roles in adhesion promotion **(Figure 1A)**.[57, 58] Mfp-1 is primarily found in the byssus cuticle, serving as a protective layer that maintains a balance between high stiffness and extensibility.[52] This property is crucial to the elasticity of the mussel byssus in response to dynamic environmental conditions.

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Additionally, mfp-2 and mfp-4 are located within the foamy interior of the byssus and are essential for linking the fibrous collagen network to the adhesive plaque.[52] Mfp-3, mfp-5, and mfp-6, present at the interface between the plaque and the substrate, enhance the final step of adhesion by enabling bond reinforcement.[52] All MFPs are characterized by a high concentration of 3,4-dihydroxyphenyl-l-alanine (DOPA), a catechol derivative of the amino acid tyrosine, which is generated through its post-translational modification. DOPA significantly contributes to the interfacial adhesion of these proteins.[57] The catechol chemistry of DOPA enables the formation of various bonds at the interface, which is a crucial aspect of its adhesive properties. The adhesive properties of catechol compounds are intrinsically dependent on redox conditions, which is a key feature of catechol chemistry. Under acidic conditions, catechol compounds like DOPA predominantly exist in a highly adhesive reduced state. DOPA forms pH-dependent coordination bonds with metal ions, a process crucial for its adhesive function.[59] As the pH nears the pKa value of 9.3, DOPA is oxidized to produce DOPA-semiquinone, followed by electron loss resulting in DOPA-quinone, which markedly decreases surface adhesion.[60] In contrast to DOPA, DOPA-quinone lacks the capacity to establish permanent connections with metal ions, hence diminishing its adhesive capabilities. Recent research indicates that oxygenated saltwater possesses a favorable redox potential and a remarkable capacity to receive electrons.[60] Notably, adjusting the pH to a somewhat basic range, similar to that of seawater (≈8.0), modifies the DOPA–Fe^{3+} stoichiometry, transitioning among mono-, bis-, and tris-complexes of DOPA–Fe^{3+}, thus enhancing adhesive connections.[60] Mussel byssus cuticles are notably abundant in mfp-1 and Fe^{3+}, which enables the establishment of persistent bis- and tris-catechol-Fe^{3+} complexes resulting in the exceptional mechanical strength and extensibility of bissus threads. The catechol moiety of DOPA enables participation in several molecular interactions, such as hydrogen bonding, electrostatic

interactions, π-π stacking, cation-π interactions, hydrophobic interactions, metal coordination, and covalent bonding **(Figure 1B)**.[52, 60]

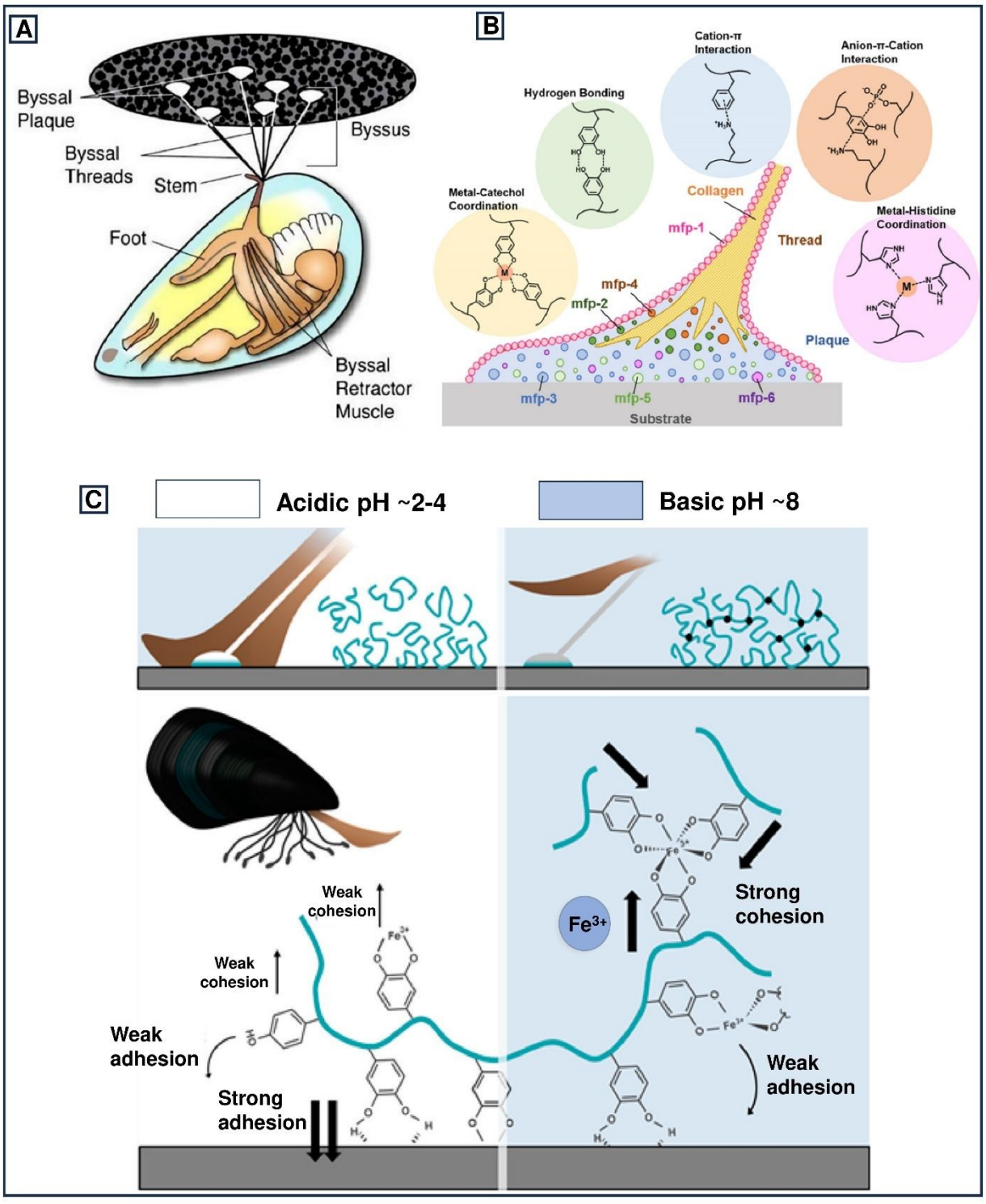

Figure 1. (A) Anatomy of the mussel and byssus structures and its attachment to the substrate. Reproduced with permission from ref.[58] under Creative Commons Attribution Noncommercial License. (B) Schematic structure of mussel byssus showing the distribution of major mussel

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foot proteins and typical reversible mussel-inspired interactions. Reproduced with permission from ref.[52] Copyright 2022, American Chemical Society. (C) Scheme showing the two-step mechanism related to the adhesion of mussel to the substrate. Reproduced with permission from ref.[61] Copyright 2016, American Chemical Society.

It is important to note that the adhesion of mussel on the substrate involves two steps including substrate adhesion and intermolecular cohesion **(Figure 1C)**.[61] First, low pH environment is created by mussel at the distal depression, followed by secretion of MFPs. MFPs, rich in DOPA, display strong adhesion to the surface. Once the strong adhesion is achieved, in the next step, this newly formed mussel byssus is exposed to sea water having Fe^{3+} and pH ~8. At this high pH, DOPA–Fe^{3+} interactions are favored resulting in strong cohesion through intermolecular bridging.

Mussels have developed ways to sustain elevated adhesion levels by adjusting the local pH surrounding their adhesive structures by ways not fully established by research. This localized acidity may retard the oxidation of DOPA, preserving it in a highly sticky state. Mussels release cysteine-rich mfp-6 at the interface when confronted with stressful conditions, promoting the reduction of DOPA-quinone back to its original DOPA form by the oxidation of the thiol group in mfp-6.[62] While mfp-6 can partially revert DOPA to its reduced form, it does not entirely prevent the oxidation of DOPA to DOPA-quinone. Nonetheless, this constraint does not diminish the overall adhesion, as oxidized DOPA may still establish covalent cross-links with other DOPA molecules, hence enhancing the cohesion and mechanical integrity of the byssus. This reversible oxidation mechanism is crucial for mussels to sustain superior adhesion in dynamic and difficult aquatic settings.

3. Catechol-based functionalization of biopolymers

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3.1. Direct addition of catechol-derivatives to biopolymer matrix

Catechol and its derivatives contain hydroxyl groups and benzene ring, which can participate in hydrogen bonding, π-π interactions, and cation-π interactions with the polymeric matrix, resulting in their uniform distribution via physical crosslinking.[63] Taking benefit of such interactions, Liao *et al.* fabricated corn starch/quaternary ammonium functionalized corn starch-based films incorporated with polydopamine (PDA).[64] Here, hydrogen bonding interactions between –OH groups of both PDA and starch helped in strengthening the developed films. These films showed good antioxidant activity (≈56.7%), and antimicrobial activity against *E. coli* (≈96.4%) and *S. aureus* (≈90%), thus extending the shelf life of strawberries by 7 days. In a similar effort to address the hydrophilicity and poor mechanical strength of pure starch films, Xu *et al.* included PDA into the films. It was observed that involvement of PDA changed the microstructure of the film and a higher water contact angle (>90°) was obtained.[65] Additionally, flexibility of the films was improved by incorporating PDA as indicated by 11 times higher elongation at break of PDA-starch films as compared to pure starch films. Next, Prabhakar *et al.* prepared PDA and ε-polylysine (ε-PL) loaded polyvinyl alcohol (PVA) based biodegradable films.[8] It was reported that including PDA in films offered reduced solubility, enhanced UV shielding properties and mechanical strength, while simultaneously providing cleavage sites for microbial degradation.

Taking a step further, Mu *et al.* developed trilayer films based on chitosan (CS), konjac glucomannan (KGM), and tragacanth gum (TG).[25] In trilayer film, physically entangled KGM and TG formed the middle layer (KT), while chitosan (CS) in combination with ε-polylysine (ε-PL) and tannic acid (TA) was used to form top and bottom layer (ECT). Here, top and bottom layer exhibited cationic-π interaction between $–NH_2$ groups of CS and ε-PL with benzene ring of TA **(Figure 2)**.[25] Further, interaction of outer layers with the middle layer was owing to the electrostatic interaction of cationic groups of outer layers with the anionic groups of TG

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(Figure 2).[25] Developed mussel-inspired trilayer films exhibited excellent barrier (2.15 g mm/m^2 day kPa), UV blocking (≈100% absorbance at 200-375 nm), and antimicrobial properties against *E. coli* (14.3 mm inhibition zone) and *S. aureus* (15 mm inhibition zone), rendering them as a suitable candidate for packaging of dry food items such as crackers.

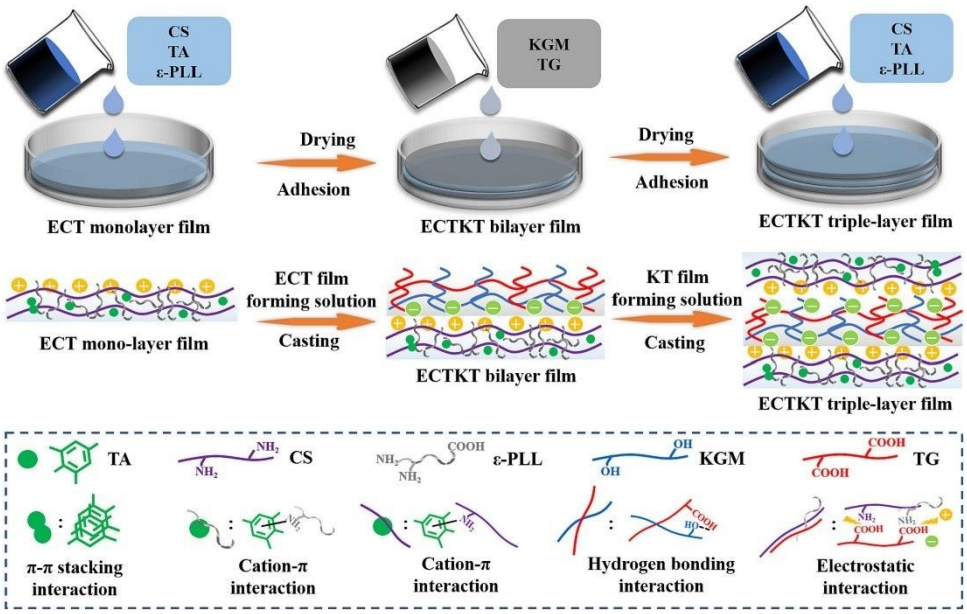

Figure 2. Schematic representation for the fabrication of mussel-inspired triple layer film and the interactions between different components. Reproduced with permission from ref.[25] Copyright 2023, Elsevier.

Next, Erihemu *et al.* prepared sodium alginate (SA) based coating solution which was crosslinked using Ca^{2+}.[66] Here, PDA was synthesized via oxidative self-polymerization of dopamine under basic conditions, and added to this coating solution which could well integrate owing to the hydrogen bonding interactions between SA and PDA. The developed SA-PDA formulation, with improved oxygen and water vapor barrier properties, was coated on potato tubers preventing their greening during 48 h storage as determined by the chlorophyll content. Further, Yu *et al.* coated cellulose paper with PDA by simple immersion method followed by dipping it in a solution containing halloysite nanotubes, polydimethylsiloxane, and stearic

acid.[67] The resulting modified paper was superhydrophobic (contact angle ≈157.5°) which could scavenge ethylene, thus extending the shelf life of cherry tomatoes.

TA, containing five catechol and five gallol groups, can easily participate in multiple interactions including hydrogen bonding, π-π interaction, and coordination bonding leading to improved properties of the developed films. Considering this, Yuan *et al.* prepared Schiff-base crosslinking-based films using dialdehyde glucomannan and gelatin.[68] TA was added during the film formation which further improved the mechanical strength of the film owing to hydrogen bonding. Apart from mechanical strength, addition of TA further provided thermal stability, UV shielding property, and antioxidant, and barrier properties to the films for their use in food packaging. Further, Yu *et al.* designed TA loaded CS/gelatin-based film where hydrogen bonding based interactions between TA and polymeric network led to ≈36% increase in mechanical strength.[69] Additionally, ≈100% UV absorbance and ≈76.5% antioxidant activity was reported for the developed films which could be used for packing premature fruits. Similarly, Zhang *et al.* utilized TA for preservation of fresh cut apples by loading it in CS/gelatin film in different concentration.[70] It was revealed that incorporating 0.5 wt% TA in films could help in blocking most of the UV light, while 2 wt% TA offered the highest antioxidant activity (≈89%). There was no significant difference reported in the capability of 1 wt% and 2 wt% TA loaded films for preserving fresh cut apples.

Further, exploring other catechol derivatives, Nilsuwan *et al.* included gallic acid (GA) or TA in chicken protein isolate (CPI)/gelatin-based films.[24] In general, incorporation of these catechol derivatives reduced the solubility of the films while improving the mechanical properties. It was reported that TA loaded films displayed lower water vapor permeability as compared to GA loaded films. However, GA loaded films had better antioxidant activity, thus reducing lipid oxidation of chicken skin oil over 15 days of storage.

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Moving ahead, to include both active and intelligent features in packaging films, Luzi *et al.* developed PVA based films incorporated with catechol derivatives namely GA or quercetin (QC).[71] A comparative study of GA and QC incorporated PVA films revealed that GA could be easily dispersed and its inclusion gave better antioxidant activity, however, the films became rigid. On the other hand, QC loaded films were flexible which may be attributed to the sole presence of –OH groups only. It was reported that the changes in color for both GA (greenish tone) and QC (reddish tone) loaded films after a specific time period, can be helpful for intelligent food packaging.

3.2. Biopolymers grafted/chemically cross-linked with catechol derivatives

Taking inspiration from Mussel, scientists have also chemically functionalized various biopolymers with catechol derivatives.[72, 73] Utilization of these functionalized biopolymers has led to the development of packaging films with remarkable physicochemical properties for food packaging application. For example, Zhao *et al.* utilized oxidized TA having quinone groups to react with $–NH_2$ groups of chitosan by forming Schiff-base based crosslinking points.[74] TA grafted chitosan could be easily combined with corn starch to make bilayer film with strong interface, owing to hydrogen bonding interactions between –OH groups of TA and starch. The film displayed ≈95% antioxidant activity with almost three times reduction in water vapor permeability (WVP) and 4.6 times increment in tensile strength. The bilayer film could prolong the shelf life of bananas by 6 days while reducing the weight loss by 14%.[74] Similarly, Lee *et al*. utilized TA based chemical crosslinking for preparing chitosan-based films.[75] Here, the crosslinking was performed at two different pH i.e., pH 7.4 and pH 8.5. It was reported that Schiff-base based crosslinking was predominant at pH 7.4 (CS-TA/P), while Michael addition-based crosslinking was more prevailing at pH 8.5 (CS-TA/T) **(Figure 3)**.[75] Experimental studies further suggested higher crosslinking density and barrier properties in the case of films

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prepared at pH 8.5 as compared to pH 7.4. Food preservation studies on banana clearly presented the better performance of chitosan films crosslinked with TA at pH 8.5, showing reduced weight loss over the period of 7 days.

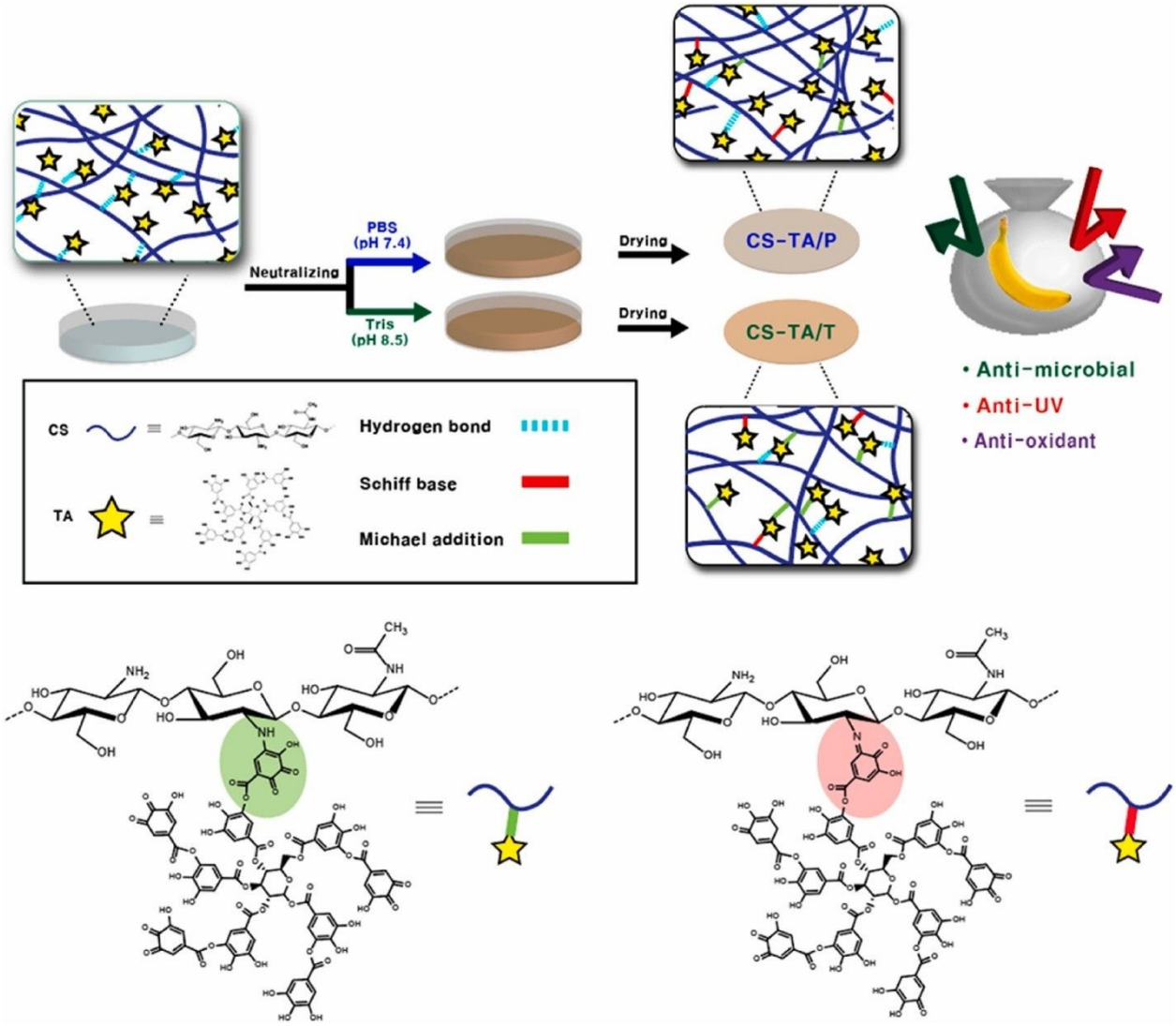

Figure 3. Scheme showing preparation of CS-TA based films at pH 7.4 (CS-TA/P) and pH 8.5 (CS-TA/T) with prevailing crosslinking mechanisms. Reproduced with permission from ref.[75] Copyright 2022, Elsevier.

Further, employing carbodiimide chemistry, catechol grafted chitosan (C-CS) was synthesized by Lei *et al.* using dihydroxyhydrocinnamic acid.[23] Next, C-CS was mixed with PVA in different ratios and films were prepared by solvent evaporation method. A significant

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increase in glass transition temperature of the films clearly indicated the presence of strong interface between C-CS and PVA. Grafting of catechol on CS helped to reduce UV transmission by ≈67% and increase the mechanical strength by ≈46% as compared to pure PVA films. Moving further, Lei *et al.* explored C-CS and PVA based matrix in combination with TA/Ti coated layered clay (LDHs@TA-Ti/C-CS/PVA).[76] It was reported that the addition of LDHs@TA-Ti in C-CS/PVA films further improved mechanical strength by ≈45% and barrier properties by ≈36%.

Next, Wang *et al.* grafted four different catechins including epicatechin (EC), epicatechin gallate (ECG), epigallocatechin (EGC), and epigallocatechin gallate (EGCG) on chitosan chains via radical mechanism.[77] Characteristic signals of respective catechin were visible in all four grafted chitosan as evidenced by nuclear magnetic resonance (NMR) spectroscopy. It was reported that the grafting of ECG and EGCG was more efficient as compared to EC and EGC. Next, these EC-CS, ECG-CS, EGC-CS, and EGCG-CS were individually mixed with pure CS to make catechin functionalized films. Among all the films, ECG-CS/CS film was most efficient in slowing down lipid oxidation in corn oil during storage owing to its superior antioxidant and barrier properties as compared to pure chitosan films.

Exploring other biopolymers, Yu *et al.* synthesized dopamine grafted carboxymethyl cellulose (CMC) which was then reacted with chitosan employing Schiff-base and Michael addition-based crosslinking.[78] This strategy could be utilized for modifying the surface of paper with layer-by-layer approach. Further, Islam *et al.* functionalized cellulose paper with succinic acid, which in turn was reacted with dopamine via carbodiimide chemistry.[79] Dopamine functionalized cellulose paper could successfully immobilize silver ions and facilitated their reduction, resulting in silver nanoparticles (Ag NPs) (50-60 nm size) decorated paper exhibiting antimicrobial properties with very limited leaching.

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In another direction of functionalizing proteins with catechol derivatives for packaging application, Lv *et al.* grafted PDA on lysozyme utilizing Schiff-base based reaction.[80] PDA-lysozyme was coated on the surface of polyethylene (PE) films by simple immersion method. These PDA-lysozyme/PE films with desired physicochemical properties could be successfully employed for maintaining the freshness of pork for 10 days. Further, Wang *et al.* functionalized soy protein with catechol groups (SPI-CH) to impart it with adhesive properties.[81] SPI-CH was mixed with soybean polysaccharide for making films which exhibited good interfacial interaction and barrier properties for food packaging application.

3.3. Catechol functionalized nanomaterials

With the advancement in nanotechnology, numerous nanomaterials have been synthesized and explored for various applications including packaging. These nanomaterials are added into the polymer matrix as a filler to include active properties into the nanocomposite while simultaneously imparting mechanical robustness and maintaining light weight feature. To achieve desired results, uniform distribution of nanomaterials inside the polymer matrix combined with good interfacial interaction is crucial.[82] To address this, utilizing the wet adhesive feature of mussel for various surfaces, scientists have functionalized different nanomaterials with catechol and its derivatives.[83] Although such mussel inspired nanocomposites have been widely used for biomedical applications, their use in packaging area has been rather limited.[84]

It has been reported that silver in its different nano forms is prone to aggregation upon mixing in polymeric matrix, which influences the overall antimicrobial properties of the developed films.[85] To solve this issue, Zhou *et al.* functionalized silver nanosheets with polydopamine (PDA@Ag) which was synthesized by oxidative self-polymerization of dopamine under basic conditions.[86] Developed PDA@Ag nanosheets exhibited good

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dispersibility, and interfacial interaction with pectin upon incorporation into pectin-based films resulting in enhanced water vapor and oxygen barrier properties. Moreover, the adhesive nature of polydopamine reduced the leaching of silver, thus offering long-term antimicrobial activity. The presence of PDA as a coating on silver nanosheets also offered good biocompatibility, UV resistance, and mechanical properties, thus helping in increasing the shelf life of mushrooms. Similarly, to improve the interfacial interaction between the polymer matrix and the nanofiller, Lodhi *et al.* functionalized cellulose nanocrystals (CNC) with PDA.[87] At first, coating of PDA was performed for different time period to optimize a thin coating on the surface of CNC (PDA@CNC). The developed PDA@CNC were mixed with carboxymethyl cellulose (CMC) to fabricate active packaging films. These films displayed better mechanical strength, thermal stability, UV resistance, water vapor barrier properties, structural integrity under wet conditions, and antioxidant activity, thus rendering them as a potential alternative for active food packaging.

It is noteworthy that poly(lactic acid) (PLA) based packaging lacks sufficient gas and UV barrier, and they do not have any antimicrobial activity.[88] Considering the importance of above-mentioned properties for packaging application, Xu *et al.* explored polydopamine coated CNC (PDA@CNC) as a filler, and PDA@CNC/PLA composite films were prepared by a combination of solution precipitation and hot pressing.[89] It was reported that including PDA@CNC in films improved their mechanical strength, and offered UV shielding feature to PLA films.

In the similar direction of imparting PLA films with additional properties, Mao *et al.* used Cu^{2+} mediated catechol coating on layered clay which was subsequently incorporated into catechol grafted chitosan/polyvinyl alcohol coating solution.[90] This was combined with PLA films to develop multilayer composite films for active food packaging. Similarly, Lei *et al.* developed tannic acid/Ti^{4+} coated layered clay which was homogeneously mixed in catechol

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grafted chitosan/polyvinyl alcohol matrix for making films displaying good gas and UV barrier properties along with antimicrobial activity.[76]

Additionally, García-Arroyo *et al.* functionalized covalent organic framework with dopamine (COF_{DOPA}) utilizing Cu mediated azide-alkyne chemistry.[91] COF_{DOPA} was uniformly distributed inside PLA as a nanofiller, resulting in reinforced PLA-based active packaging films with non-migratory antioxidant component.

Further, Li *et al.* prepared PDA coated graphene oxide (PGO) and silver NPs (Ag NPs).[92] PGO was prepared by simple self-polymerization of dopamine on the surface of GO under basic conditions **(Figure 4)**. Ag NPs were prepared by reduction of silver salt with the help of PDA, combined with strong affinity of PDA for metal offering stability to the synthesized Ag NPs. PGO and Ag NPs were then added to a solution of gelatin/cellulose nanofibrils (CNF), and the films were prepared by solvent casting method. These thermally stable and UV resistant films displayed uniform distribution of the nanomaterials in the polymer matrix owing to good interfacial interaction offered by catechol groups **(Figure 4)**.[92] In a similar direction, Li *et al.* prepared dopamine grafted CNF which displayed interfacial assembly with montmorillonite clay.[93] The resulting nanohybrids were mixed in gelatin-based matrix where catechol groups were taken into account for *in situ* reduction of silver salt, leading to the formation of antimicrobial films for active food packaging.

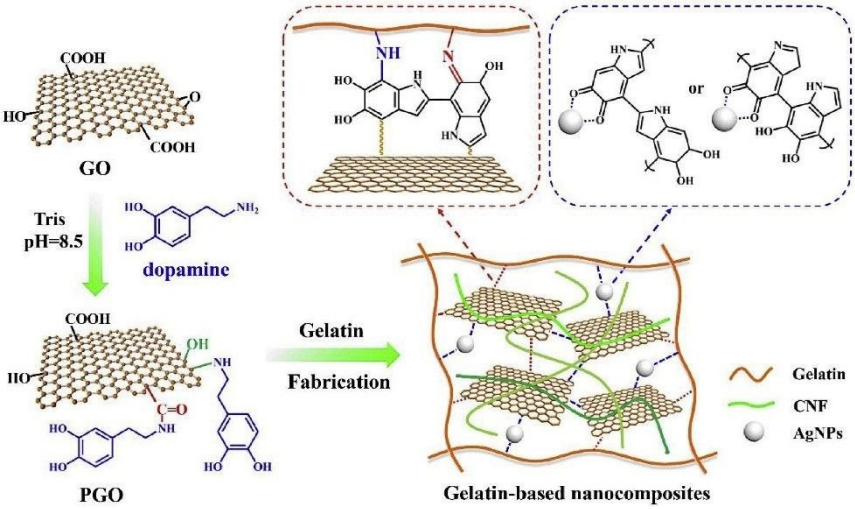

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Figure 4. Development of PGO nanohybrids and its utilization in forming gelatin/cellulose nanofibrils based films loaded with Ag NPs. Reproduced with permission from ref.[92] Copyright 2019, Elsevier.

4. Mussel-inspired coatings for food preservation

Active packaging materials with antioxidant and antimicrobial properties facilitate in protecting the food item from microbial spoilage and other environmental factors. Such materials can be either used as a packaging film or can be applied as a coating on the surface of fruits and vegetables. Surface of the food item can be coated with the active packaging solution by various methods including dip/immersion coating, spraying, brush coating, and layer by layer coating with subsequent air drying **(Figure 5A)**.[94, 95] Dip/immersion coating is one of the most commonly used methods on laboratory scale owing to its simplicity and low cost, however, it requires large drying space and more time for evaporating water used as a solvent.[96] Brushing method helps to avoid the immersion of food items into the solution, however, it is less efficient. Spraying method involves the atomization of liquid into droplets which in turn spread over the surface of the food item.[97] This method is efficient, generates uniform coating, and avoids wastage of solvent, making it suitable for industrial application. Further, food preservation performance of the coatings can be adjusted by controlling their structure and functional properties, which offer protection from various environmental factors such as oxygen, moisture, and microorganisms **(Figure 5B)**.[94]

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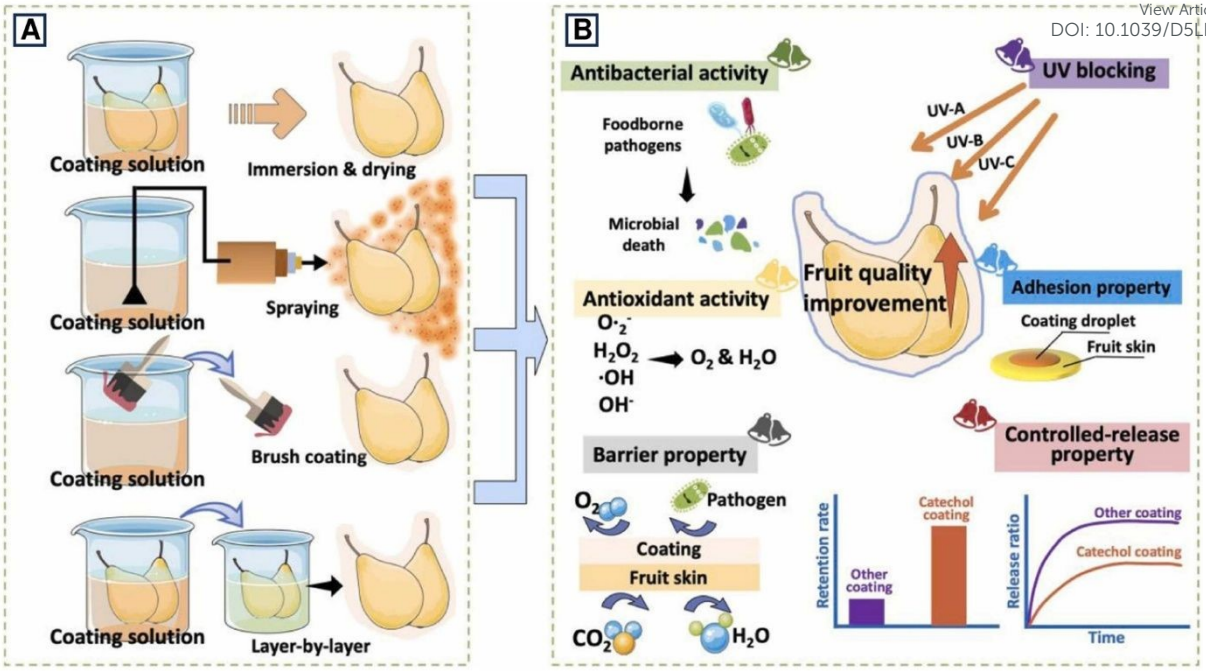

Figure 5. (A) Schematic representation of different methods used for coating the food items. (B) Improvement of the shelf-life of food items by offering various physicochemical and active properties. Reproduced with permission from ref.[94] Copyright 2025, Elsevier.

For effective coverage of the surface and formation of a uniform thin film, the adhesion of the coating solution on the target surface is very critical. It is governed by the surface energy and the wettability, which affect the spreading capability of the solution on the surface of the food item.[98] Mussel-inspired coating formulation provides good adhesion between coating and food surface, thus allowing the formation of a uniform coating which can effectively improve the shelf-life of the food item **(Figure 6A)**.[94] On the contrary, non-adhesive coating formulations can slide off the surface quickly due to the lack of sufficient adhesion, thus leaving patches of the food surface exposed to the environmental factors, which may ultimately lead to faster food spoilage **(Figure 6B)**.[94] Considering this, mussel-inspired catechol-based chemistry has very recently emerged as a technological solution for developing adhesive coatings for active food packaging.

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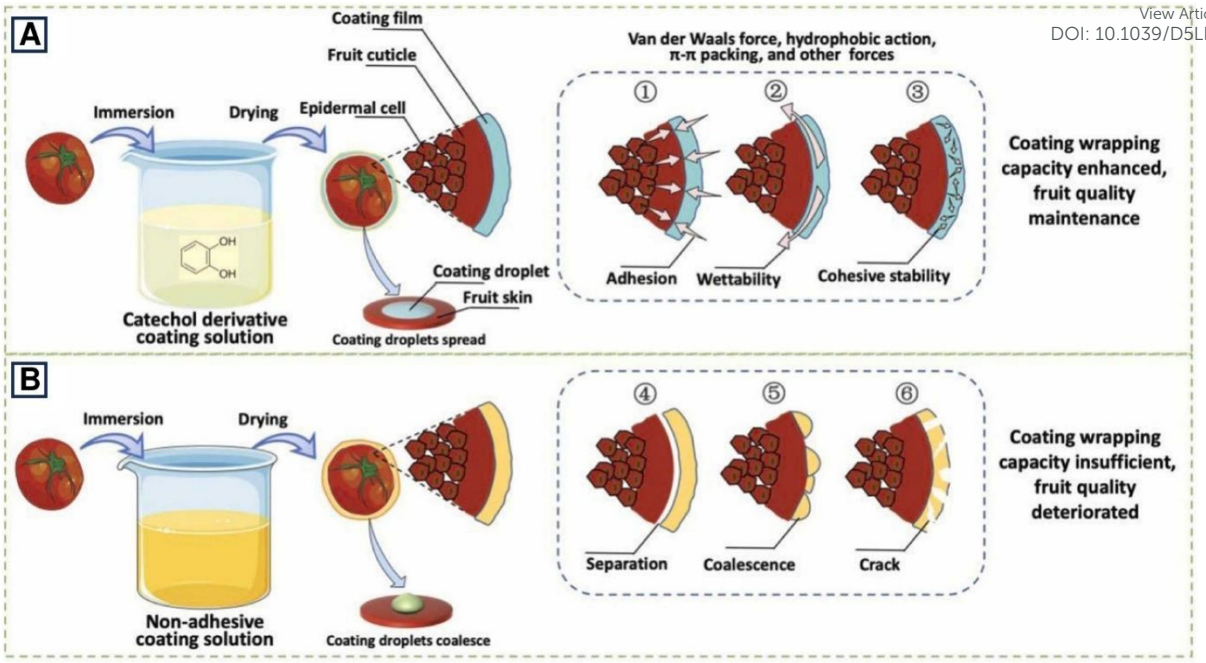

Figure 6. Comparison of the adhesive properties of coatings prepared by using (A) catechol derivative containing coating solution, and (B) non-adhesive coating solution, and their impact on food quality. Reproduced with permission from ref.[94] Copyright 2025, Elsevier.

In this direction, He *et al.* synthesized tannic acid functionalized chitosan having quaternized amine groups (QCS-TA).[99] Further, crosslinking of QCS-TA with oxidized chitosan (OCS) by Schiff-base based crosslinking approach offered the possibility of making both active packaging film and coating **(Figure 7)**.[99] Based on the incorporated catechol groups of tannic acid and quaternized amine groups, the developed films and coatings showed excellent antioxidant, UV resistance, and antimicrobial properties. Schiff-base based crosslinking and catechol groups-based hydrogen bonding helped in imparting good tensile strength (≈70 MPa), water vapor barrier (≈14 $g \cdot h^{-1} \cdot m^{-2}$), and oxygen barrier (≈3.5 $g \cdot mm \cdot m^{-2} \cdot h^{-1}$) properties. Shear thinning behavior of developed formulation indicated its potential for making a thin film (≈3 μm thickness) on the surface of fruits and vegetables. The antioxidant activity of the formulation was ≈97%, and it showed ≈98% antimicrobial activity against both *E. coli* and *S. aureus*. Here, positively charged formulation could interact with negatively

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charged phospholipid membrane of bacteria causing membrane disruption resulting in bacterial cell death. Further, 90% cell viability against L929 cells confirmed the biocompatibility of the developed system. Additionally, no adverse effect was observed on the growth of bean sprouts further indicating the non-toxic nature of the formulation. In the next step, food preservation studies were performed by coating the formulation on fresh strawberries and bananas. The experimental results revealed the capability of the coatings in reducing weight loss, preventing bacterial growth, maintaining color, and extending the shelf -life of strawberries and bananas by 6 and 5 days, respectively **(Figure 8)**.[99] Additionally, this nanocomposite formulation was also used as active packaging films to pack mushrooms where the shelf life was improved by 6 days, thus showing dual purpose coating/packaging film potential for food preservation.

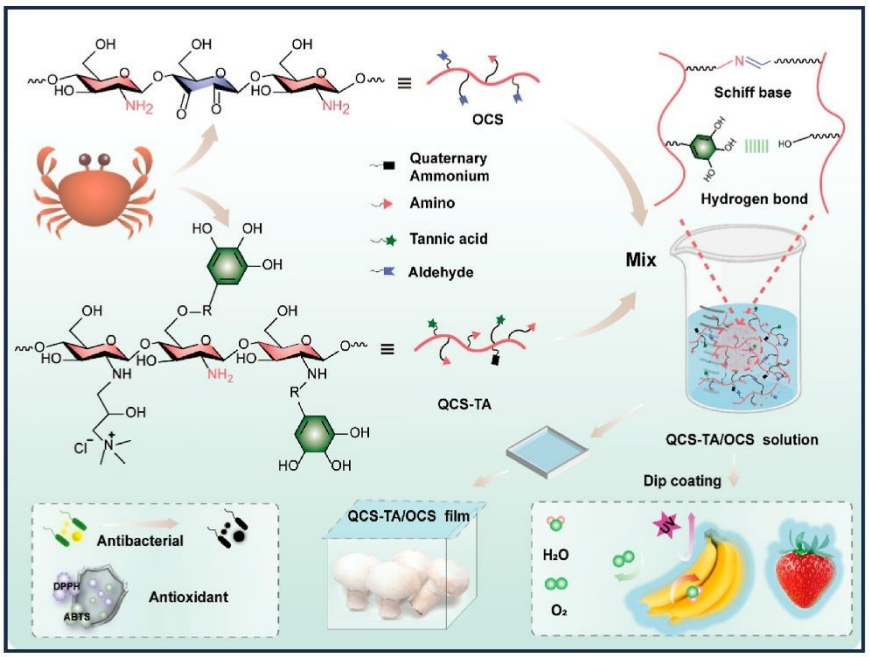

Figure 7. Schematic representation of the fabrication process of quaternary ammonium and tannic acid modified chitosan/oxidized chitosan (QCS-TA/OCS) coating and QCS-TA/OCS film for fruit preservation. Reproduced with permission from ref.[99] Copyright 2024, American Chemical Society.

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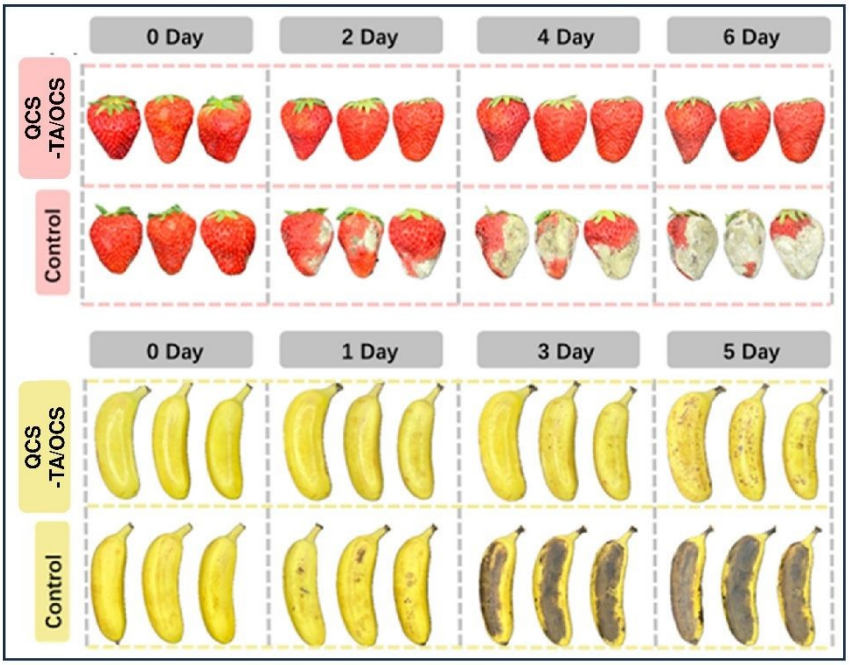

Figure 8. Images displaying impact of QCS-TA/OCS coating on the ripening of strawberries and bananas. Adapted with permission from ref.[99] Copyright 2024, American Chemical Society.

Similarly, Zhou *et al.* developed mussel-inspired adhesive coating using catechol-functionalized quaternized chitosan (CQ-CS) that was prepared by grafting 2, 3-epoxypropyl trimethyl ammonium chloride and 3, 4-dihydroxy benzaldehyde on chitosan polymer chains **(Figure 9A)**.[100] CQ-CS based coatings had excellent surface adhesion combined with excellent mechanical, antioxidant and antimicrobial properties which helped in food preservation **(Figure 9B)**.[100] Strawberry and banana were used as model fruits here, and coating was simply performed via dip coating for 5 s using 1 wt% CQ-CS solution followed by storage at 25 °C and 75% relative humidity. Bare strawberries and strawberries coated with quaternized chitosan (QCS) were used as controls for comparison. Interestingly, a simple visual inspection of strawberries with different treatments over seven days indicated the potential of CQ-CS for keeping strawberries fresh for a longer period of time **(Figure 10)**.[100] Here, mussel-inspired CQ-CS based adhesive coating could reduce the weight loss in strawberries, and helped in

maintaining the color which could otherwise change post-harvesting due to the degradation of pigments. Additionally, CQ-CS coatings could keep the strawberries fresh indicated by lower degree of variation in total soluble solids which provides the information related to the delayed degradation of polysaccharides and ripening of fruits. The lowest titratable acidity was observed in the case of CQ-CS coating indicating its potential in lowering the respiratory progress. Further, CQ-CS coatings were also tested on bananas where the appearance of black spots on the skin could be delayed up to 10th day, thus extending its shelf life.

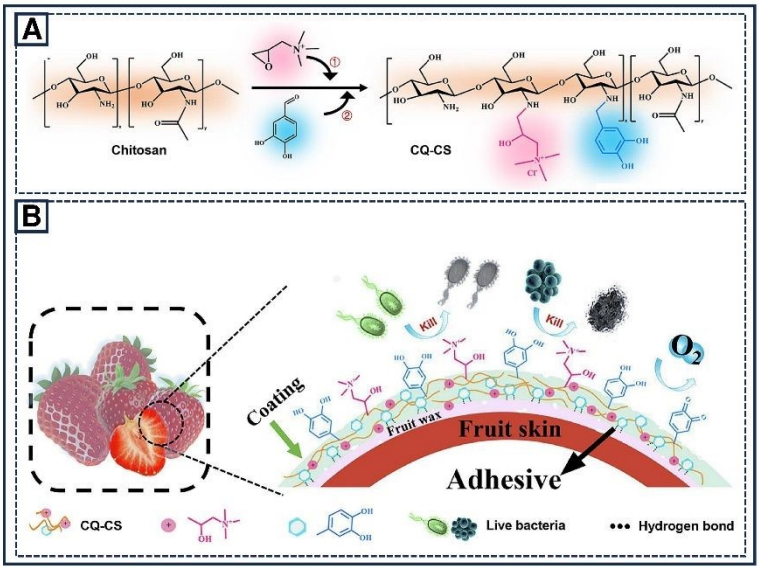

Figure 9. (A) Schematic representation for the preparation of catechol-functionalized quaternized chitosan (CQ-CS). (B) Coating of strawberries with CQ-CS for killing the bacteria and keeping them fresh during storage. Adapted with permission from ref.[100] Copyright 2023, Elsevier.

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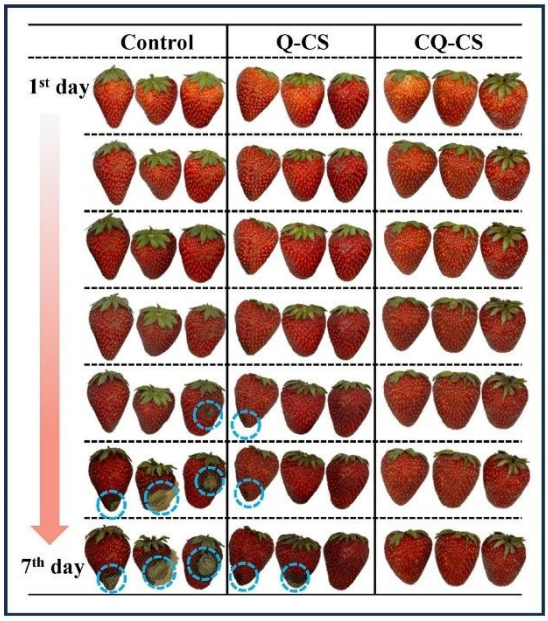

Figure 10. Digital images showing changes in QCS and CQ-CS coated strawberries over 7 days. Untreated strawberries were used as control. Reproduced with permission from ref.[100] Copyright 2023, Elsevier.

To improve the adhesion on hydrophobic surface of fruits and vegetables, Huang *et al.* grafted epigallocatechin gallate (EGCG) on low methoxy pectin (LMP) by free radical mechanism.[101] Simple dip coating method was used for 60 s to coat LMP-EGCG on grapes. Contact angle measurements exhibited good wettability and adhesion of LMP-EGCG coatings on grapes. This coating displayed ≈80% antioxidant activity, and ≈85% and ≈87% antimicrobial activity against *E. coli* and *S. aureus*, respectively. Further, fruit preservation studies revealed that LMP-EGCG coatings could help in reducing the weight loss, lipid oxidation, and growth of microbes. It also reduced the activity of polyphenol oxidase which oxidizes polyphenols, thus slowing down the browning of grapes.

In another direction, Park *et al.* developed spray-based nano-coatings that can be applied on different surfaces and could be used for large scale applicability.[102] Here, concept of supramolecular chemistry was utilized to develop Fe^{3+}/tannic acid-based coating which

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could be coated on any surface by spraying for less than 5 s. This antioxidant and antimicrobial spray coating was further evaluated for improving the shelf life of oranges and strawberries. Preservation study performed for 14 days using coated and uncoated oranges clearly showed that coated oranges maintained their integrity up to 14 days while uncoated oranges were spoiled with visible growth of greenish mold **(Figure 11A)**.[102] Next, oranges were sprayed with the coating solution using commercially available spraying machine. It was reported that none of the coated oranges was spoiled even after 28 days while 27% of bare oranges were spoiled **(Figure 11B)**.[102] In a separate study, strawberries on the trees were sprayed with the coating solution for less than 3 s followed by spraying of fresh tap water in the evening. These coated strawberries were plucked next day and stored in a box. Uncoated strawberries were used as a control here and almost all of them were rotten after 58 h of storage **(Figure 11C, D)**.[102] In contrast, more than half of the coated strawberries still remained edible, thus rendering the spray-based coating as a promising alternative **(Figure 11C, D)**.[102]

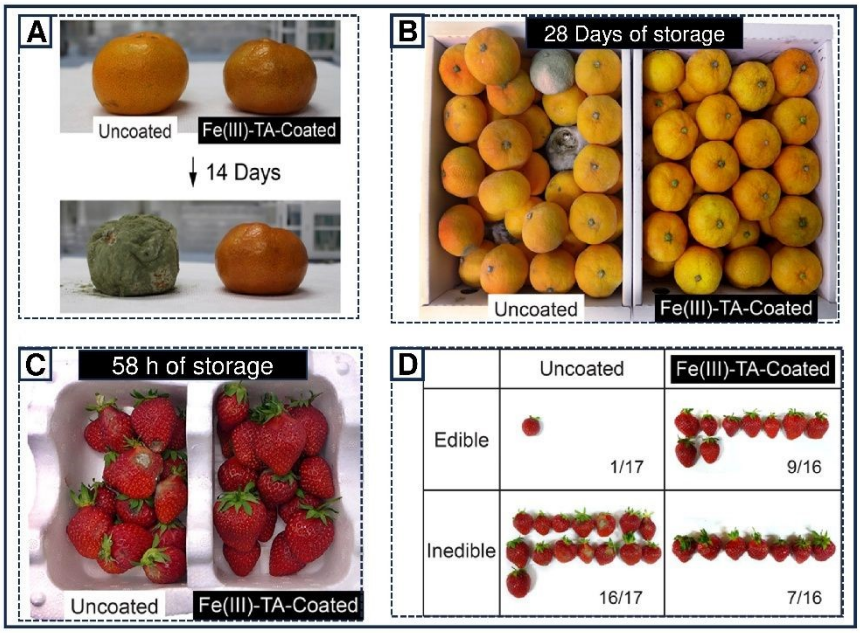

Figure 11. (A) Digital images of uncoated and Fe(III)-TA-coated mandarin oranges after storage at room temperature over 14 days. (B) Visual changes in mandarin oranges coated by commercially available spraying machine and its comparison with uncoated mandarin oranges

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over 28 days. (C) Visual changes in coated and uncoated strawberries, and (D) statistical analysis in terms of edible and nonedible strawberries. Adapted with permission from ref.[102] under Creative Commons Attribution (CC BY) license.

Considering hydrogen bonding and electrostatic interactions exhibited by mussel utilizing catechol and amino acids, Li *et al.* fabricated adhesive coatings using curcumin-zein-riboflavin-carrageenan (CZRC) nanocomposite.[103] Here, the capability of carrageenan to bind with water and biogenic amine via hydrogen bonding and electrostatic interactions helped in achieving good adhesion. This was also supplemented by zein which is a hydrophobic material containing amino acids, thus helping in adhering to meat surface upon the evaporation of solvent.[103, 104] Further, added curcumin and riboflavin not only imparted antioxidant activity, but also acted as photosensitizers to disrupt bacterial cell membrane and offer light triggered sterilization. Here, the treatment of CZRC with blue light for 30 min provided excellent antimicrobial efficacy against *Pseudomonas fragi* and *Brochothrix thermosphacta* to protect the packed lamb and pork from bacterial contamination. It was reported that CZRC coating with blue light treatment significantly lowered the total volatile basic nitrogen, delayed change in pH, and reduced lipid oxidation for both lamb and pork. Overall, this approach could extend the shelf life of lamb and pork by 1.8 and 2.3 times, respectively.

In addition to tannic acid, polyphenols containing propolis extract (PPE) has also been explored for making edible coatings. The composition of the PPE differs based on the geographical location.[105] It is expected that the catechol derivatives present in these polyphenols can help in developing coatings on fruits while simultaneously providing active properties. In this direction, Moreno *et al.* developed gelatin/PPE based edible coating for protecting raspberries from fungal infection.[106] Here, two different approaches were used for incorporating PPE (a) direct incorporation in gelatin, and (b) encapsulation of PPE in zein

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nanocapsules which were integrated in gelatin. The developed coating exhibited broad spectrum antifungal activity with highest efficiency against *P. digitatum* and *B. cinerea*. It was reported that the encapsulation of PPE was a better approach for prolonging its release, thus ensuring long term protection for raspberries.

In another approach, taking inspiration from remarkable adhesion of naturally occurring mussels, Yu *et al.* synthesized dopamine grafted carboxymethyl cellulose (CMC-g-DA).[78] In the next step, CMC-g-DA and chitosan were coated on a paper using layer by layer technique followed by oxidative crosslinking using sodium periodate. Modified paper exhibited excellent mechanical properties and water vapor barrier properties. Also, it exhibited more than 90% antimicrobial activity against both *E. coli* and *S. aureus*. This modified paper could effectively be used for keeping the mushrooms fresh for 6 days.

Further, Xie *et al.* reported lignin/tannin/ZnO nanoparticles-based adhesive coating formulation for developing modified paper with good heat and UV resistance, and antimicrobial activity combined with lower water vapour permeability.[107] Modified paper exhibited excellent mechanical strength owing to the fact that biomimetic coating not only covered the surface, but also penetrated inside the spaces between the fibers, thus strongly binding them together. Similarly, Roman *et al.* explored catechol containing polyphenols for modifying polypropylene (PP) films, imparting them with antioxidant activity.[108] Further, Mao *et al.* used catechol containing quercetin for coating layered double hydroxide which were subsequently mixed with catechol grafted chitosan/polyvinyl alcohol solution. This adhesive nanocomposite solution was coated on the surface of polylactic acid (PLA) films to provide them with excellent antimicrobial and UV barrier properties for potential food packaging application.[90] Although the above-mentioned modified paper, PP and PLA based films displayed multiple functionalities, they still need to be evaluated for their real-life applicability.

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The recent examples of mussel inspired catechol-based coatings along with a comparison of their characteristic features is given in **Table 1**.

Table 1. Recent examples of mussel inspired catechol-based coatings for food packaging and a comparative analysis of their key performance parameters.

SN	Matrix	Water vapor barrier (transmission rate/permeability)	UV-barrier	Antioxidant activity (%)	Antimicrobial activity against	Coating method	Food item for Packaging test	Reference
1	QCS-TA/OCS	≈14.3 $g \cdot h^{-1}$ m^{-2}	Present	ABTS (≈97); DPPH (≈45)	*S. aureus* and *E. coli*	Brush coating	Strawberries and bananas	He *et al.*[99]
2	CQ-CS	–	–	ABTS (≈90); DPPH (≈85)	*S. aureus* and *E. coli*	Dip coating	Strawberries and bananas	Zhou *et al.*[100]
3	LMP-EGCG	–	–	DPPH (≈80)	*S. aureus* and *E. coli*	Dip coating	Grapes	Huang *et al.*[101]
4	CZRC	–	–	DPPH (≈89); ABTS (≈ 91)	*P. fragi* and *B. thermosphacta*	Spray	Meat preservation (lamb and pork)	Li *et al.*[103]
5	lignin/tannin/ZnO nanoparticles	–	Present	–	*S. aureus* and *E. coli*	Brush coating on paper	–	Xie *et al.*[107]
6	Polyphenols @PP	–	Present	–	*E. coli*	–	–	Roman *et al.*[108]

7	CMC-g-DA/chitosan	≈1.3 ×10^{-10} g m/ Pa·s·m^2	–	–	*S. aureus* and *E. coli*	Dip coating on paper	Mushrooms	Yu *et al.*[78]
8	Fe^{3+}/tannic acid	–	Present	–	–	Spray coating	Oranges and strawberries	Park *et al.*[102]

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5. Safety and sustainability of mussel-inspired coatings

Mussel-inspired coatings, predominantly those based on catechol chemistry, are promising options for developing sustainable, high-performance food packaging or coating formulations due to their excellent adhesion in humid environments and compatibility with biodegradable polymer matrices. However, addressing both safety and sustainability is vital for their widespread implementation. Public and regulatory bodies have continuously scrutinized composite packaging materials, raising concerns about increasing consumer exposure to potential toxic additive components in packaging resulting from their migration into food.[109] In this context, the European regulations 1935/2004/EC and 450/2009/EC require that any active or intelligent packaging elements must preserve or improve food quality without releasing harmful substances.[109] Consequently, determining the leaching and migration of packaging components during contact with food simulants or actual food is essential.

Biopolymers derived from natural sources—such as starch, chitosan, cellulose, and proteins—are biodegradable, edible, and generally regarded as safe for food preservation. Their integration with catechol-based compounds raises new safety considerations. Dopamine, a common catechol used to mimic mussel adhesion, can pose potential risks. It is a neurotransmitter and it possesses the ability to generate reactive oxygen species (ROS) or reactive metabolites upon its dysregulation.[110] Due to these traits, it not only can potentially hinder with the normal cell signaling but also can damage cellular components.[110] Although polydopamine (PDA) nanoparticles have shown biocompatibility in vitro and in vivo, there is limited understanding of their metabolic degradation pathways and the long-term fate of any breakdown products.[111] Furthermore, other studies claim that a high-dose exposure to mesoporous PDA (at 78.57 mg/kg) in animal models has been associated with gut microbiota disruption, altered bile acid and fatty acid metabolism, inflammation pathway activation, and

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oxidative stress.[112] Therefore, for dopamine-functionalized food packaging films in direct contact with food, understanding dopamine migration and exposure dosage is crucial.

Safety assessments currently include cytotoxicity studies and animal feeding trials. For example, HeLa cell viability above 90% was observed when cultured with starch-based films containing 0.5 wt% PDA nanoparticles. In a 30-day mouse gavage study, no organ lesions or adverse weight effects were observed, demonstrating acceptable safety in vivo **(Figure 12)**.[64] Other studies indicate that dopamine functionalization on other carrier particles may reduce their migration rate. For instance, PLA films incorporating dopamine-modified antioxidant covalent-organic frameworks (COFs) exhibited total migration well below the EU regulatory limit of 60 mg/kg in food simulants, owing to dopamine-induced cross-linking that halted COF migration.[91] However, the research on the toxicity of dopaminated composites is still lacking.

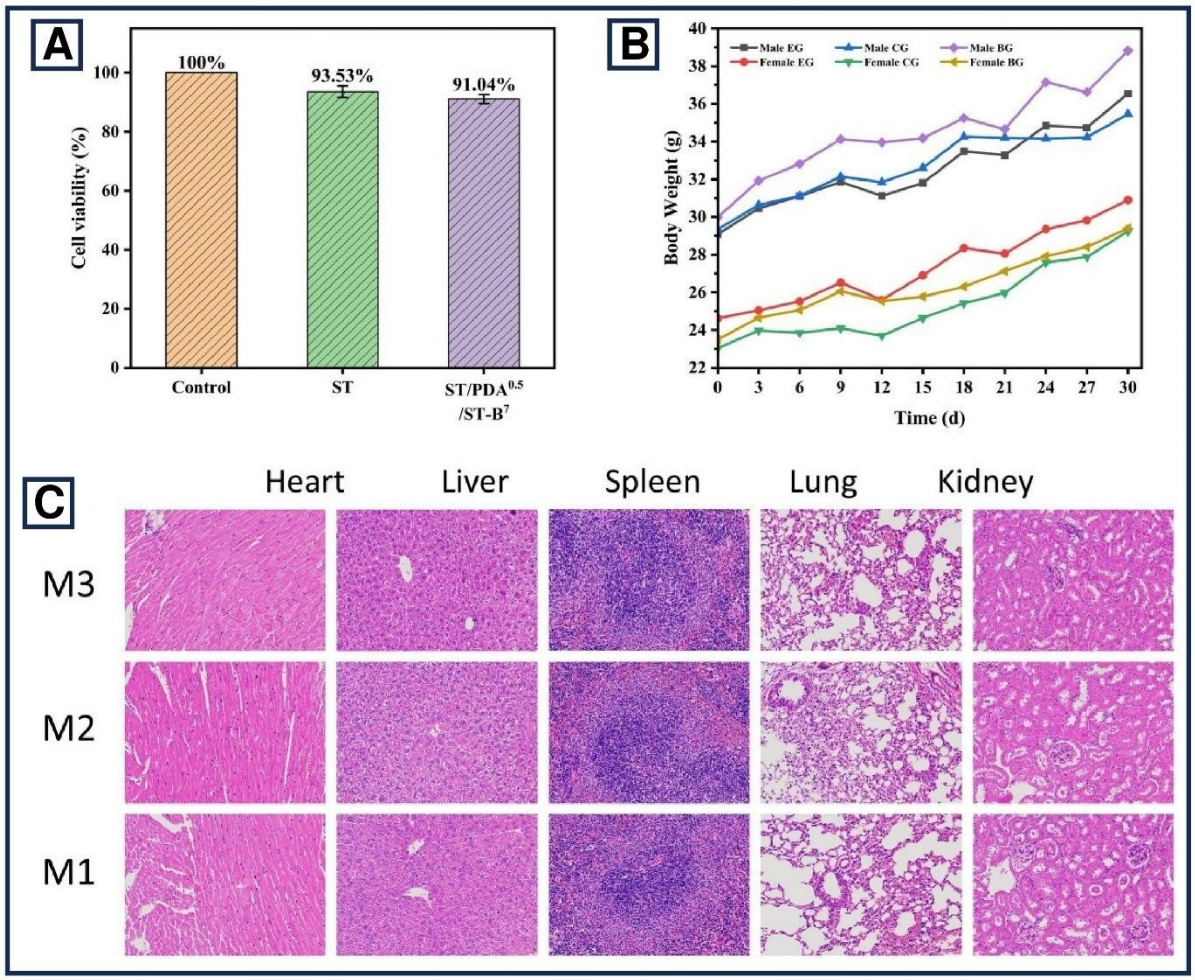

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Figure 12. (A) Effect on the viability of HeLa cells when cultured with neat cornstarch films (ST) and films comprising 93% cornstarch, 3% rosin quaternary ammonium salt-modified cornstarch (ST-B) and 0.5 wt% PDA (ST/PDA$^{0.5}$/ST-B^{7}) for 3 days. (B) changes in body weight of male and female mice gavaged with film solutions over a 30 day period. (C) H&E staining of major organs in male mice 30 days after treatment with ST/PDA$^{0.5}$/ST-B^{7} showing no signs of abnormalities. Adapted with permission from ref.[64] Copyright 2024, Elsevier.

While safety issues regarding the use of dopamine persist, plant-derived catechol derivatives such as tannic acid (TA) are generally recognized as safe (GRAS) when used within regulated limits. Although TA exhibits toxicity at high concentrations,[113] food packaging films typically use TA at 1-5 wt%, with migration levels into simulants substantially lower than dietary intake from fruits and vegetables. Casein films containing up to 10 wt% TA showed no cytotoxicity, while higher concentrations (>10 wt%) elicited cell stress, primarily due to unbound TA.[114] Encouragingly, washing the films significantly reduced toxicity, and TA migration into food was effectively absent. Nonetheless, the safety of certain modified catechols may also depend on the toxicity profiles of residual synthesis reagents. For instance, acylated tannins used in starch-nanofiber films contained residual DMSO (4.07 mg/m²).[115] Despite its compliance with permissible non-benzene solvent limits (<5 mg/m²), the degradation products and chemical residues must be carefully evaluated for food contact guidelines.

Edible coatings are subjected to stricter ingestion standards compared to films. Typically, plant-based catechols are more popular in developing edible coatings and dopamine is generally excluded. Edible coatings are often designed to be washed off before consumption or applied to inedible peel layers.[116, 117] Washable coatings and peel barriers effectively limit additive ingestion. However, since mussel-inspired coatings are designed to be water resistant,

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the edibility of the additives is inevitable along with the food. Therefore, toxic compounds must be excluded from being used, and other GRAS compounds must be used within the safe limits. Nonetheless, further investigations into molecular transport behavior within coating matrices and across biological barriers are warranted.

From a sustainability standpoint, mussel-inspired coatings must align with circular economy principles—utilizing renewable feedstocks, low-toxicity processing, and enabling biodegradability and recyclability. Biobased polymer matrices offer substantial environmental benefits, including reduced carbon footprints and improved end-of-life outcomes compared to fossil-derived plastics. However, relying solely on biodegradable polymers does not guarantee sustainability. Infrastructure limitations, potential microplastic formation, additive-induced ecotoxicity, and consumer confusion with labels can undermine environmental goals. Mussel-inspired coatings must therefore incorporate green solvent systems—such as water-ethanol blends for PDA deposition—to minimize volatile organic compound emissions. Key sustainability indicators include post-use degradability and recyclability. Catechol-functionalized composites based on cellulose or starch matrices have shown favorable biodegradation profiles,[118] aligning with open environmental regulations. However, biopolymer-based formulations are rarely intended for end-of-life recycling or reprocessing, and biodegradation is their only fate.

6. Conclusions and future perspectives

Biopolymer-based food packaging films are being extensively developed as sustainable substitutes for petroleum-based plastic food packaging films. Current research endeavors concentrate on enhancing and adjusting the characteristics of biopolymer-based food packaging films. Mussel biomimetic approaches have emerged as an intriguing and effective strategy for enhancing the performance of these films. The functionalization of biopolymer-

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based food packaging films with catechol derivatives, including DA and TA, has attracted considerable interest over the last decade. This method demonstrates significant potential in enhancing the mechanical strength, barrier properties, and functional attributes of biopolymer-based food packaging films. Catechol derivatives inspired by mussels can be incorporated into packaging films using various methods, utilizing their capacity for multiple physical and chemical interactions. The resultant composite films demonstrate a diverse range of functionalities, encompassing robust interfacial adhesion, photothermal properties, UV absorption, and free radical scavenging capabilities. These attributes allow them to fulfil various functions within packaging matrices. Moreover, the improved adhesion afforded by edible coatings derived from mussel-inspired techniques augments the interfacial interaction between the coating solution and the food surface, thereby amplifying the preservation efficacy of the edible coating. This study emphasizes that biopolymer-based food packaging films and coatings, developed using mussel-inspired methods, demonstrate favourable safety profiles and are appropriate for food-contact applications.

Future challenges persist in the advancement of biopolymer-based food packaging films functionalized with catechol derivatives through mussel-inspired methodologies. The polymerization and cross-linking mechanisms of catechol derivatives, including DA and TA, in various biopolymer solutions are not well understood. The molecular mechanism of direct polymerization, along with the influence of pH, ionic strength, and temperature on cross-linking behavior and resultant film properties, necessitates additional research. Future research must concentrate on elucidating the cross-linking mechanisms between catechol derivatives and biopolymers comprehensively, while precisely delineating the correlation among cross-linking mechanisms, film structure, and performance.

Moreover, a comprehensive evaluation of the safety and migration properties of catechol derivatives in biopolymer-based food packaging films is necessary. The

polymerization or modification of catechol derivatives in these films may change their safety profile, requiring reassessment. Future research must prioritize thorough safety evaluations to guarantee food contact safety. The distinctive characteristics of DA and TA, including photothermal activity and regulated release, offer prospects for the creation of food packaging materials with enhanced functionalities. Research should focus on developing mussel-inspired biopolymer-based food packaging films with improved and varied functional properties to effectively address contemporary food preservation requirements.

Current research on mussel-inspired edible coatings primarily targets fresh fruits and vegetables; however, future investigations should broaden their application to encompass additional food products. Special emphasis must be placed on the interfacial interaction characteristics of edible coatings with these novel substrates, encompassing adhesion and preservation efficacy. Ultimately, the economic viability of biopolymer-based food packaging films must be meticulously evaluated to expedite their development. This entails assessing the cost-effectiveness of performance enhancement strategies and formulating scalable preparation methods appropriate for extensive industrial production, rather than limited laboratory environments.

Conflicts of interest

There are no conflicts of interest to declare.

Data Availability Statement

No primary research results, software or code have been included and no new data were generated or analysed as part of this review.

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Abbreviations: ABTS = 2,2'-azino-bis (3-ethylbenzothiazoline-6-sulfonic acid); Ag NPs = Silver nanoparticles; C-CS = Catechol grafted chitosan; CMC = Carboxymethyl cellulose; CNC = Cellulose nanocrystals; CNF = Cellulose nanofibrils; CQ-CS = Catechol-functionalized quaternized chitosan; CPI = Chicken protein isolate; CS = Chitosan; CZRC = Curcumin-zein-riboflavin-carrageenan; DA = Dopamine; DHI = Dihydroxyindole; DOPA = 3,4-dihydroxy-L-phenylalanine; DPPH = 2,2-diphenyl-1-picrylhydrazyl; EC = Epicatechin; ECG = Epicatechin gallate; EGC = Epigallocatechin; EGCG = Epigallocatechin gallate; GA = Gallic acid; KGM = Konjac glucomannan; LMP = Low methoxy pectin; NE = Norepinephrine; NMR = Nuclear magnetic resonance; OCS = Oxidized chitosan; PDA = Polydopamine; PE = Polyethylene; PGO = PDA coated graphene oxide; Polyphenols@PP = Catechol containing polyphenols for modifying polypropylene based coating; PPE = Propolis extract; PVA = Polyvinyl alcohol; QCS = Quaternized chitosan; SA = Sodium alginate; SPI-CH = Soy protein with catechol groups; TA = Tannic acid; TG = Tragacanth gum; ε-P = ε-polylysine; WVP = Water vapor permeability

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No primary research results, software or code have been included and no new data were generated or analysed as part of this review.