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

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In recent years, taking inspiration from mussel, an underwater organism, various packaging alternatives including adhesive coatings have been developed for food preservation to ensure food availability for everyone. The extraordinary adhesion exhibited by mussel is mainly offered by mussel foot proteins 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 the recent efforts made for 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 the successful commercial utilization of such biomimetic coatings in the future.

1. Introduction

The world population is estimated to touch ≈9.8 billion by 2050, making food security a prime concern.[1] According to a report by the United Nations, ≈1 billion ton 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 consumed by the consumer.[3] This situation of food wastage is very critical, and it affects the economy of every country, demanding technological interventions to ensure food availability for everyone.[4]

In order to achieve the global sustainable development goal of "zero hunger" put forward by the United Nations, food packaging plays a crucial role.[5] Packaging acts as a physical barrier safeguarding the food from outside contamination.[6]

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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 wastes are generated every year by the 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 by sensing the loss of freshness over time.[10–17]

Development of packaging materials requires a polymeric matrix, which is either made up of a single polymer or prepared by blending different polymers. In this regard, natural polymers (*e.g.*, polysaccharides and proteins) have emerged as excellent choices for matrix owing to their inherent biocompatibility and biodegradability.[18] Further, the 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 nanomaterials is very crucial to achieve the desired physicochemical properties. Additionally, in the case of edible coatings, satisfactory adhesion of the coating solution onto the surface of fruits and vegetables is critical to avoid dripping and achieve uniform coverage.[21]

Naturally existing mussel is known to be an "underwater expert" owing to its very strong adhesion on variety of surfaces facing shear stress caused by flowing water.[22] This extraordinary adhesion is achieved by mussel foot proteins 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 the packaging sector for improving the shelf life of food items.[23–26]

In the last decade, a plethora of studies dealing with different aspects of food packaging materials have been reported in the literature.[27–32] Moreover, 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 its 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 the recent advancement in the utilization of mussel-inspired catechol-based coatings for food preservation while 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

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 media 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)

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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 synthesized, which have unique physicochemical attributes (Scheme 1). DOPA, the most common derivative, acts as a precursor of DA in humans (Scheme 1A and B). The presence of a –COOH group at the R1 position is the characteristic feature of DOPA, and it is naturally synthesized by the catalytic action of tyrosine hydroxylase enzyme. Different analogues of DOPA such as L-DOPA and 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 the brain.[36] The coating of polyDOPA on substrates has been reported to be less favorable than 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 onto 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 coating conditions, the polymerization rate of NE is slower than that of PDA. Additionally, the thickness of the NE coating (≈20 nm) is lesser than that of the PDA coating (≈40 nm).[39] NE-based coatings have been successfully developed on a variety of substrates ranging from glass to polymers.[40]

The derivatization of an $–NH_2$ 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, the conjugation of catechol moieties facilitates the coating of substrates. Various substituents like 2-bromoisobutyryl, methyl, pyrrole, pyridine, and methacrylate have been utilized for developing functionalized coatings (Scheme 1D).[40,41]

The incorporation of various substituents such as $–NO_2$, –Cl, –Br, and –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, and 5-hydroxydopamine can be utilized for preparing functional coatings with varying features (Scheme 1E).[36,42–44] For example, 6-nitrodopamine is more oxidation resistant than 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-1*H*-indazole in basic media/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 been widely explored as coating materials owing to the presence of catechol groups.[47–49]

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 under wet or underwater conditions. The loss of adhesion results from the development of 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 com-

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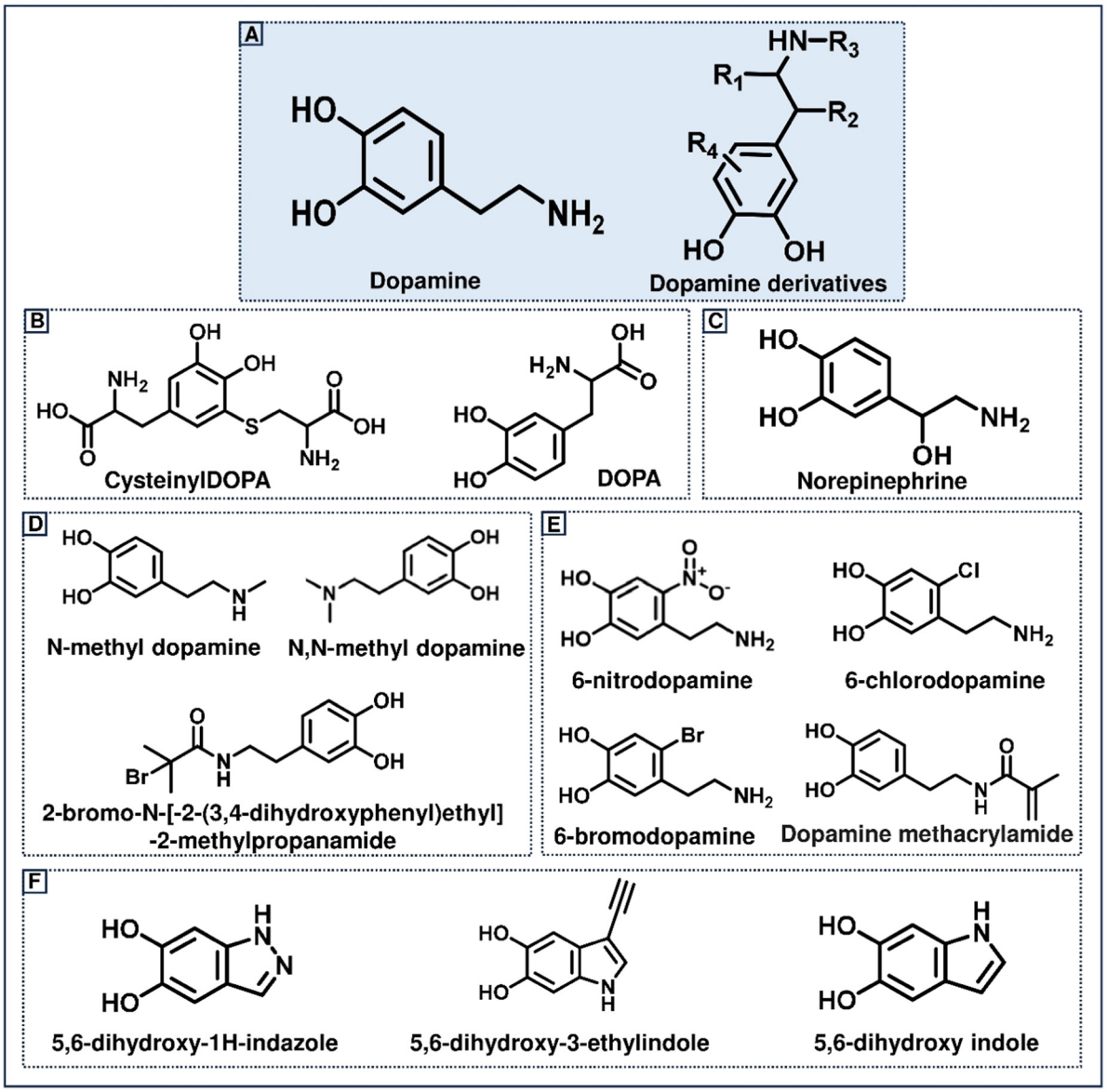

Scheme 1 (A) Chemical structures of dopamine and its derivatives prepared by chemical modifications at different sites depicted by R1, R2, R3, and R4. Functionalization at (B) R1 and (C) R2 positions of the alkyl chain, (D) R3 position having an amine group, and (E) R4 position having aromatic sites.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

pared 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 (Fig. 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. 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 cate-

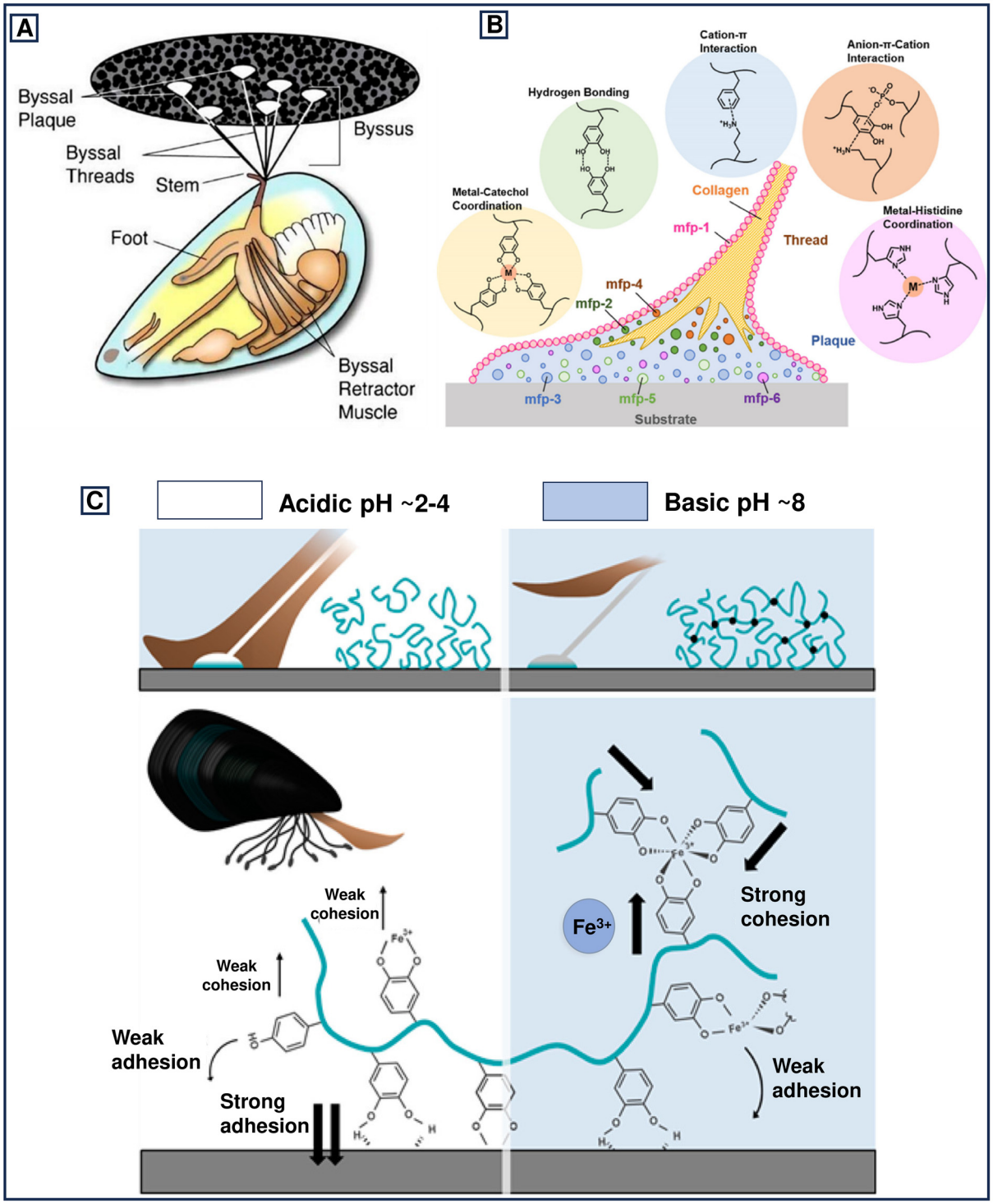

Fig. 1 (A) Anatomy of the mussel and byssus structures and its attachment to the substrate. Reproduced with permission from ref. 58 under Creative Commons Attribution Noncommercial License. (B) Schematic of the structure of mussel byssus, showing the distribution of major mussel foot proteins and typical reversible mussel-inspired interactions. Reproduced with permission from ref. 52. Copyright 2022, the 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, the American Chemical Society.

chol 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 pK_a 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 (Fig. 1B).[52,60]

It is important to note that the adhesion of mussel on the substrate involves two steps, namely, substrate adhesion and intermolecular cohesion (Fig. 1C).[61] First, a low pH environment is created by mussels at the distal depression, followed by the 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 *via* 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 crosslinks 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

3.1. Direct addition of catechol-derivatives to the biopolymer matrix

Catechol and its derivatives contain hydroxyl groups and a 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] Making use of such interactions, Liao *et al.* fabricated quaternary ammonium-functionalized corn starch-based films incorporated with polydopamine (PDA).[64] Here, hydrogen bonding interactions between the –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 the involvement of PDA changed the microstructure of the film, and a higher water contact angle (>90°) was obtained.[65] Additionally, the flexibility of the films was improved by incorporating PDA, as indicated by 11 times higher elongation at break of PDA-starch films than 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 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 films, 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 the top and bottom layers (ECT). Here, the top and bottom layers exhibited cationic–π interaction between $-NH_2$ groups of CS and ε-PL with the benzene ring of TA (Fig. 2).[25] Further, the 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 (Fig. 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), making them suitable candidates for the packaging of dry food items such as crackers.

Next, Erihemu *et al.* prepared a 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 inter-

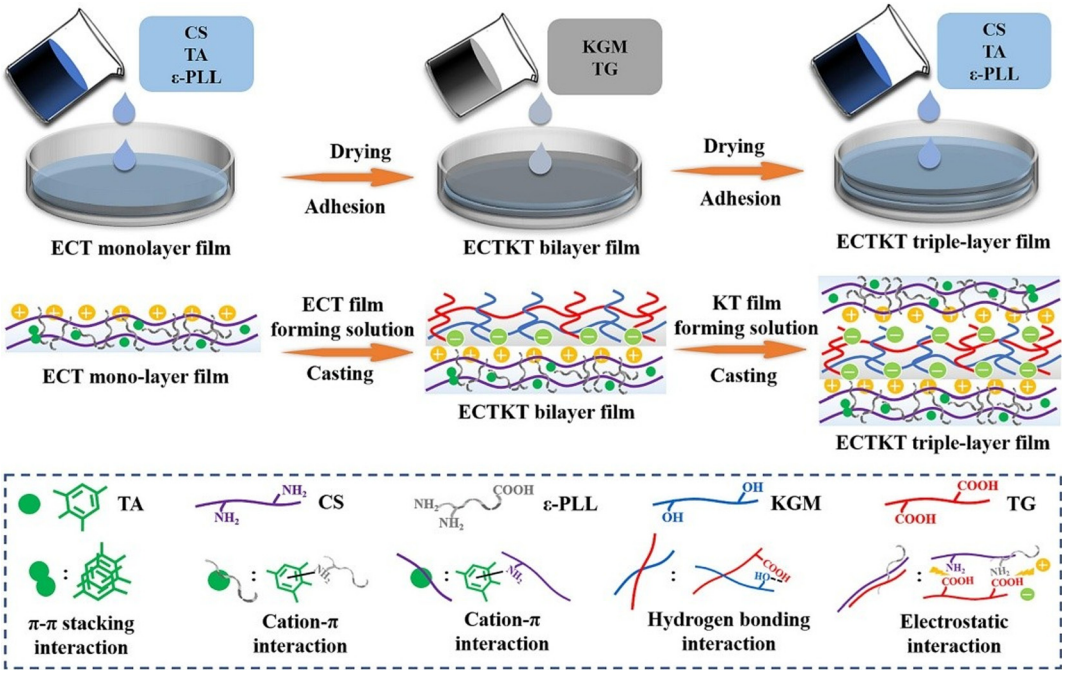

Fig. 2 Schematic of the fabrication of the mussel-inspired triple layer film and the interactions between different components. Reproduced with permission from ref. 25. Copyright 2023, Elsevier.

actions 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 a simple immersion method followed by dipping 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, the 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 the preservation of fresh cut apples by loading it in a CS/gelatin film at different concentrations.[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, the 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 the lipid oxidation of chicken skin oil over 15 days of storage.

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 activities; however, the films became rigid. However, 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] The 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 the hydrogen bonding interactions between the –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 values, *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) (Fig. 3).[75] Experimental studies further suggested higher crosslinking density and barrier properties in the case of films prepared at pH 8.5 than at 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.

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 a solvent evaporation method. A significant increase in glass transition temperature of the films clearly indicated the presence of strong interface between C-CS and PVA. The grafting of catechol on CS helped to reduce UV transmission by ≈67% and increase the mechanical strength by ≈46% as compared to

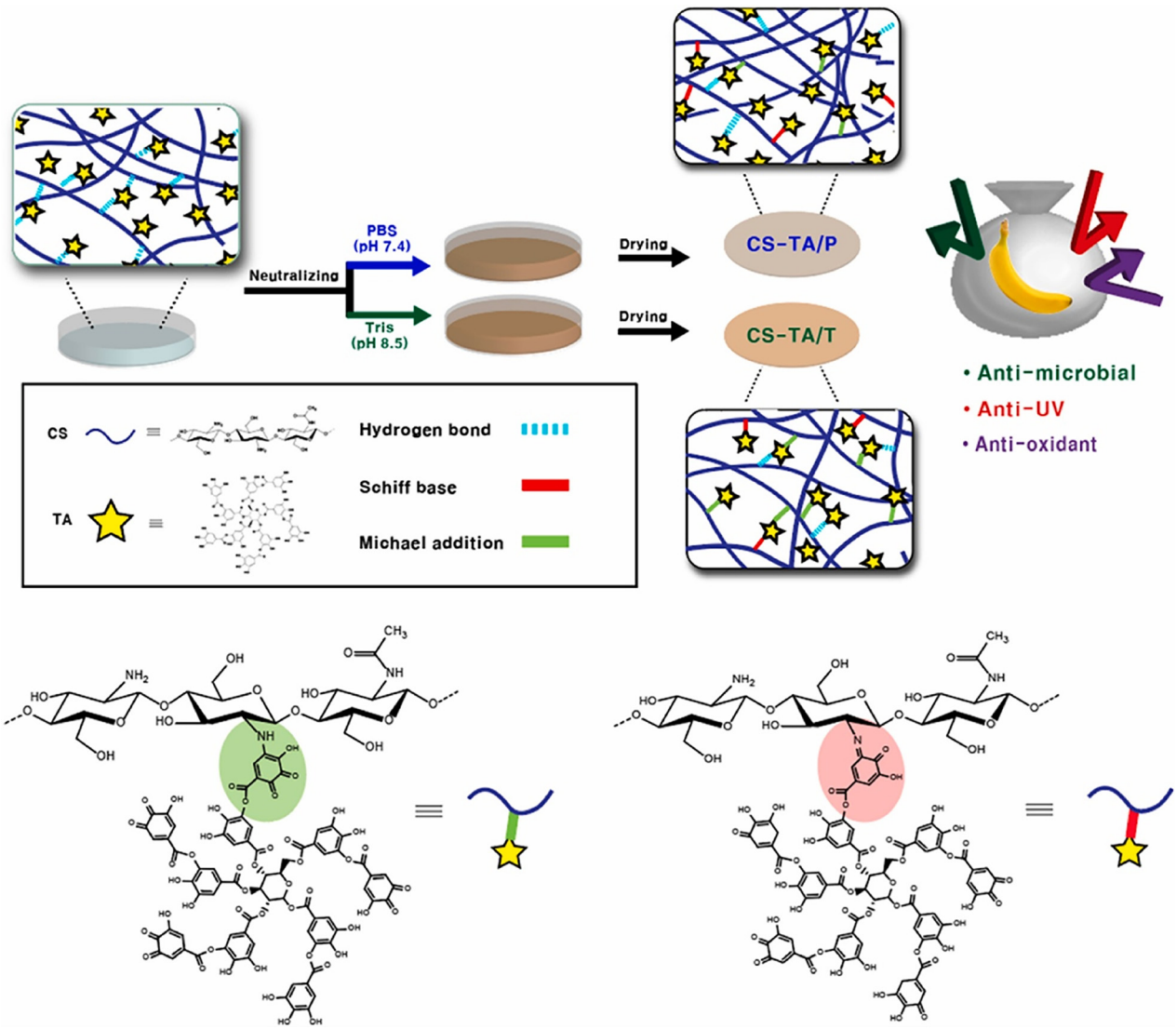

Fig. 3 Scheme showing the preparation of CS-TA-based films at pH 7.4 (CS-TA/P) and pH 8.5 (CS-TA/T) with prevailing crosslinking mechanisms. Reproduced with permission from ref. 75. Copyright 2022, Elsevier.

pure PVA films. Moving further, Lei *et al.* explored C-CS- and PVA-based matrixes 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 the 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] The 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 than that of 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 *via* a 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 facilitate their reduction, resulting in silver nanoparticle (Ag NP) (50–60 nm size)-decorated paper exhibiting antimicrobial properties with very limited leaching.

In another direction of functionalizing proteins with catechol derivatives for packaging application, Lv *et al.* grafted PDA on lysozyme utilizing Schiff-base-based reactions.[80] PDA-lysozyme was coated on the surface of polyethylene (PE) films by a 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 a 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 applications.

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 imparting mechanical robustness and maintaining light weight feature. To achieve the 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 a 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 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 (CNCs) with PDA.[87] At first, the coating of PDA was performed for different time periods to optimize a thin coating on the surface of CNC (PDA@CNC). The developed PDA@CNC was 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 making them potential alternatives 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 the above-mentioned properties for packaging applications, 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 a UV shielding feature to PLA films.

In a similar direction of endowing PLA films with additional properties, Mao *et al.* used Cu^{2+}-mediated catechol coating on layered clay, which was subsequently incorporated into a 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 a catechol-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 a 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 components.

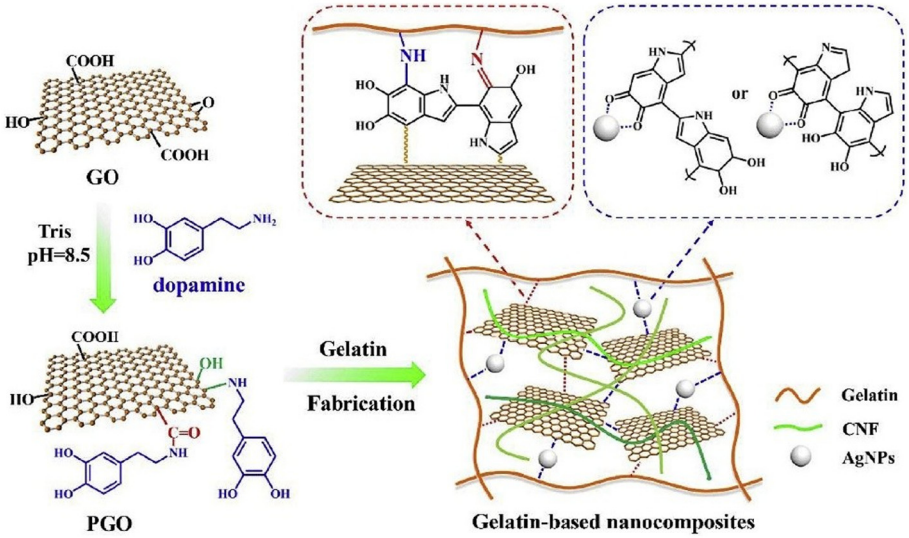

Fig. 4 Development of PGO nanohybrids and its utilization in forming gelatin/cellulose nanofibril-based films loaded with Ag NPs. Reproduced with permission from ref. 92. Copyright 2019, Elsevier.

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 (Fig. 4). Ag NPs were prepared by the reduction of silver salt with the help of PDA, combined with strong affinity of PDA for metals, 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 a 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 (Fig. 4).[92] In a similar direction, Li *et al.* prepared dopamine-grafted CNF, which displayed an interfacial assembly with montmorillonite clay.[93] The resulting nanohybrids were mixed in a gelatin-based matrix, where catechol groups were taken into account for *in situ* reduction of silver salts, leading to the formation of antimicrobial films for active food packaging.

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 applied as a coating on the surface of fruits and vegetables. The surface of the food item can be coated with an active packaging solution by various methods including dip/immersion coating, spraying, brush coating, and layer-by-layer coating with subsequent air drying (Fig. 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. The 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 applications. 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 (Fig. 5B).[94]

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 (Fig. 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 (Fig. 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.

In this direction, He *et al.* synthesized tannic acid-functionalized chitosan having quaternized amine groups (QCS-TA).[99] Further, the crosslinking of QCS-TA with oxidized chitosan (OCS) by a Schiff-base-based crosslinking approach offered the possibility of making both active packaging films and coatings (Fig. 7).[99] Based on the incorporated catechol groups of tannic

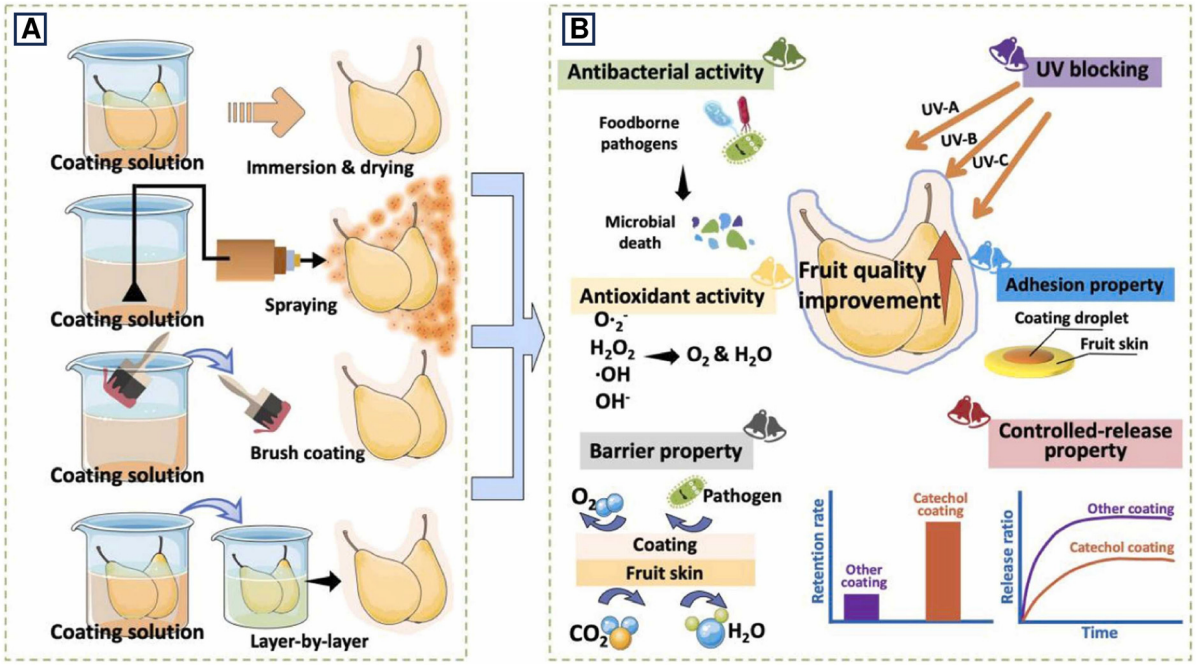

Fig. 5 (A) Schematic of different methods used for coating food items. (B) Improvement of the shelf-life of food items by offering various physico-chemical and active properties. Reproduced with permission from ref. 94. Copyright 2025, Elsevier.

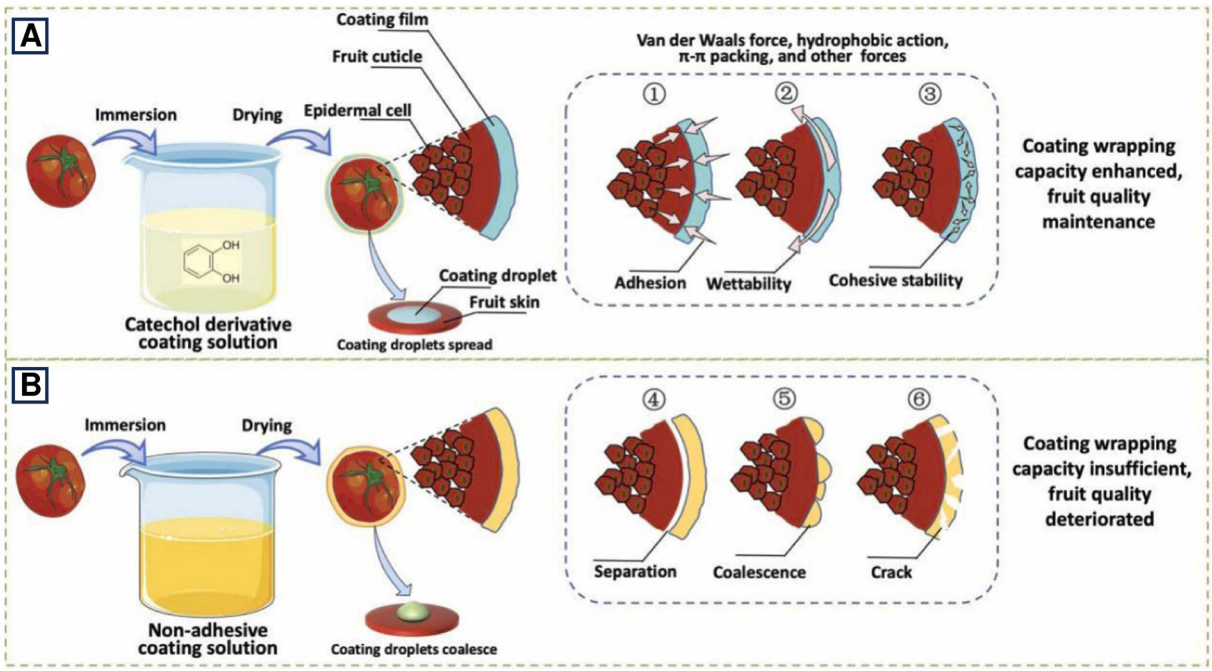

Fig. 6 Comparison of the adhesive properties of coatings prepared using (A) catechol derivative-containing coating solution and (B) non-adhesive coating solution, and their impact on food quality. Reproduced with permission from ref. 94. Copyright 2025, Elsevier.

acid and quaternized amine groups, the developed films and coatings showed excellent antioxidant, UV resistance, and antimicrobial properties. Schiff-base-based crosslinking and catechol group-based hydrogen bonding helped in imparting good tensile strength (≈70 MPa), water vapor barrier (≈14 g h^{-1} m^{-2}), and oxygen barrier (≈3.5 g mm m^{-2} h^{-1}) properties. The shear thinning behavior of the developed formulation indicated its potential for making a thin film (≈3 μm thickness) on

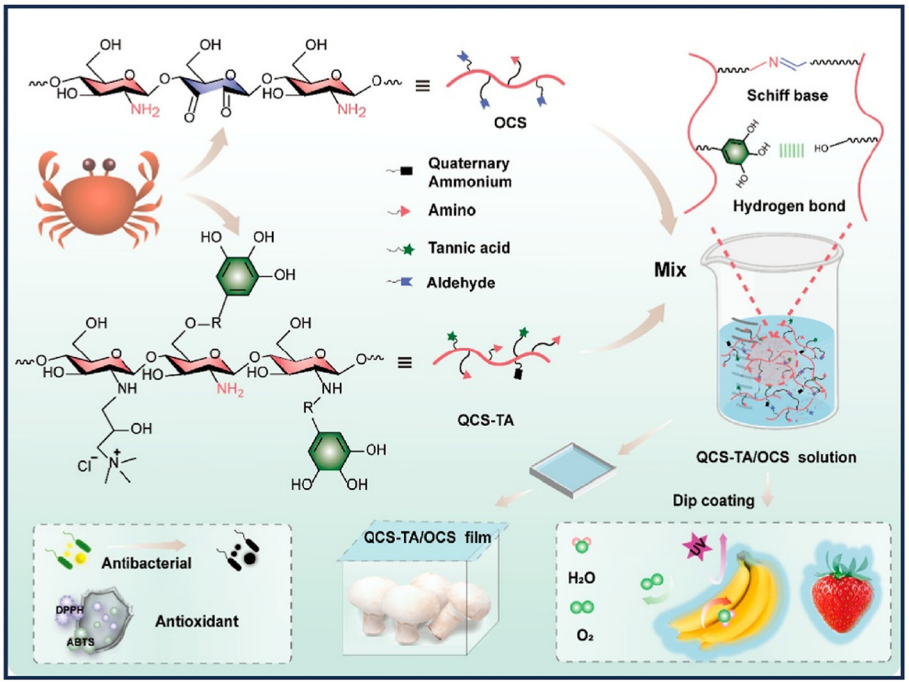

Fig. 7 Schematic of the fabrication process of quaternary ammonium- and tannic acid-modified chitosan/oxidized chitosan (QCS-TA/OCS) coating and QCS-TA/OCS film for fruit preservation. Reproduced with permission from ref. 99. Copyright 2024, the American Chemical Society.

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 the negatively 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 (Fig. 8).[99] Additionally, this nanocomposite formulation was used as an active packaging film to pack mushrooms, where the shelf life was improved by 6 days, thus showing dual purpose coating/packaging film potential for food preservation.

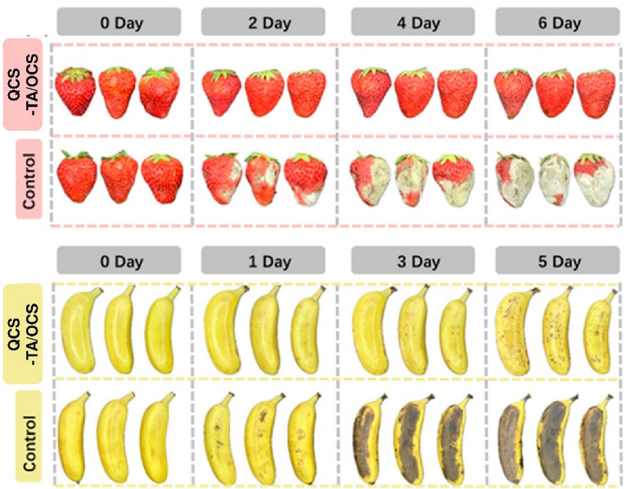

Fig. 8 Images displaying the impact of QCS-TA/OCS coating on the ripening of strawberries and bananas. Adapted with permission from ref. 99. Copyright 2024, the American Chemical Society.

Similarly, Zhou *et al.* developed a 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 (Fig. 9A).[100] CQ-CS-based coatings had excellent surface adhesion combined with excellent mechanical, antioxidant and antimicrobial properties, which helped in food preservation (Fig. 9B).[100] Strawberry and banana were used as model fruits here, and the coating was simply performed *via* dip coating for 5 s using a 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 (Fig. 10).[100] Here, mussel-inspired CQ-CS-based adhesive

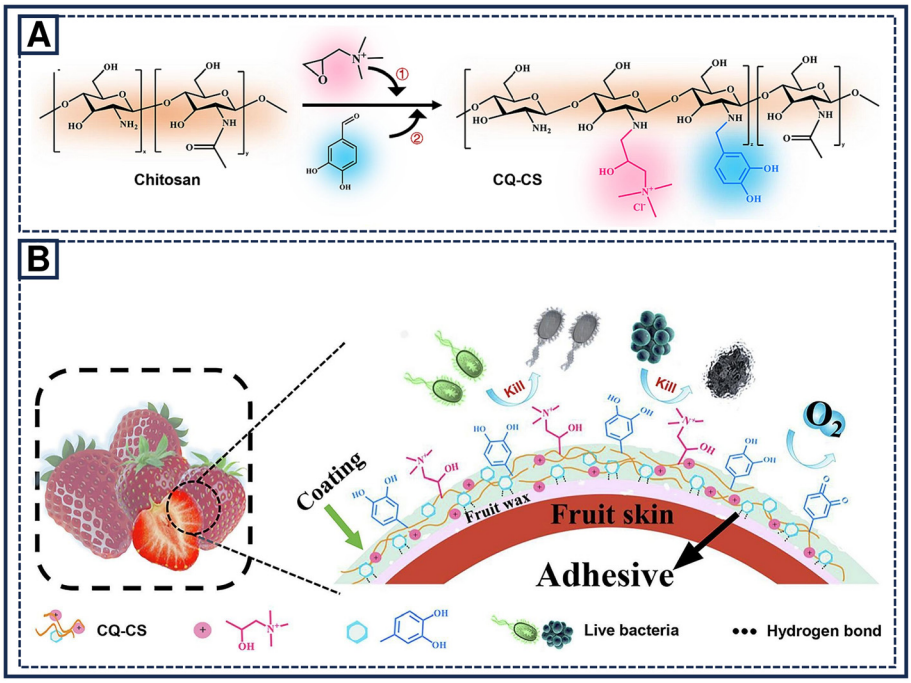

Fig. 9 (A) Schematic of the preparation of catechol-functionalized quaternized chitosan (CQ-CS). (B) Coating of strawberries with CQ-CS for killing the bacteria and keeping them fresh during storage. Adapted with permission from ref. 100. Copyright 2023, Elsevier.

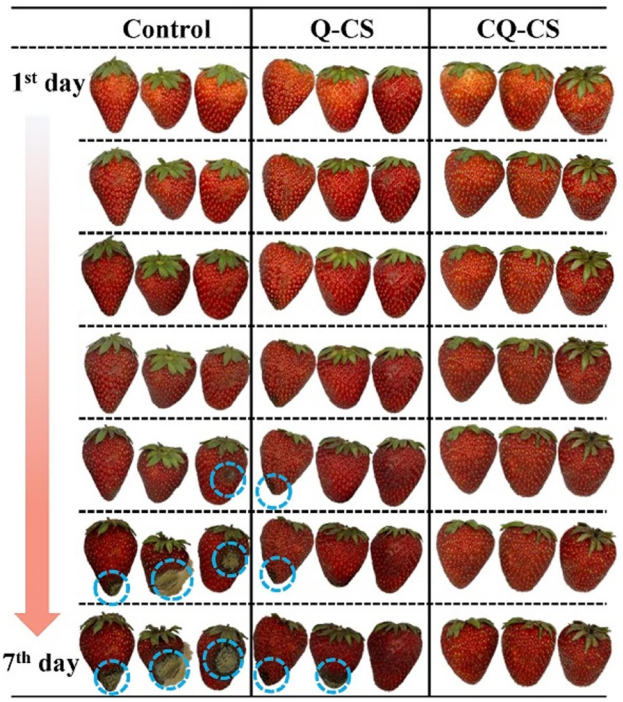

Fig. 10 Digital images showing changes in QCS- and CQ-CS-coated strawberries over 7 days. Untreated strawberries were used as control. Reproduced with permission from ref. 100. Copyright 2023, Elsevier.

coating could reduce the weight loss in strawberries and help 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 a 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 tested on bananas where the appearance of black spots on the skin could be delayed up to 10th day, thus extending its shelf life.

To improve the adhesion on the hydrophobic surface of fruits and vegetables, Huang *et al.* grafted epigallocatechin gallate (EGCG) on low methoxy pectin (LMP) by a free radical mechanism.[101] A 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 applications.[102] Here, the concept of supramolecular chemistry was utilized to develop Fe^{3+}/tannic acid-based coatings, which could be applied 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. The preservation

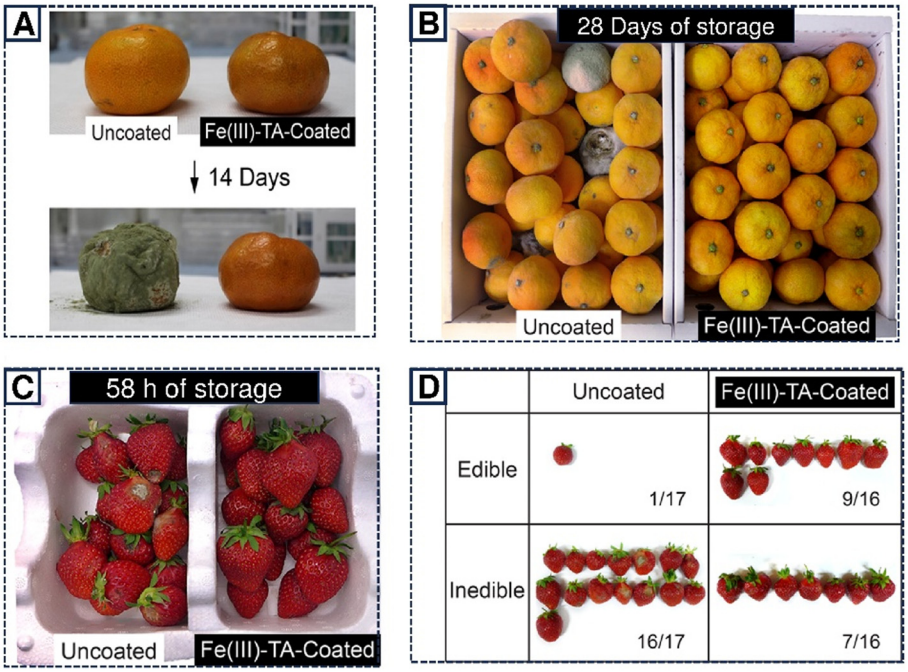

Fig. 11 (A) Digital images of uncoated and Fe(III)-TA-coated mandarin oranges after storage at room temperature for over 14 days. (B) Visual changes in mandarin oranges coated using commercially available spraying machine and its comparison with uncoated mandarin oranges for over 28 days. (C) Visual 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.

study conducted 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 (Fig. 11A).[102] Next, oranges were sprayed with the coating solution using a 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 (Fig. 11B).[102] In a separate study, strawberries on the trees were sprayed with a 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 (Fig. 11C and D).[102] In contrast, more than half of the coated strawberries still remained edible, thus making the spray-based coating a promising alternative (Fig. 11C and D).[102]

Considering hydrogen bonding and electrostatic interactions exhibited by mussel utilizing catechol and amino acids, Li *et al.* fabricated adhesive coatings using a 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 solvents.[103,104] Further, added curcumin and riboflavin not only imparted antioxidant activity but also acted as photosensitizers to disrupt the 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, polyphenol-containing propolis extract (PPE) has 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 providing active properties. In this direction, Moreno *et al.* developed gelatin/PPE-based edible coatings 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 nanocapsules, which were integrated in gelatin. The developed coating exhibited broad-spectrum antifungal activities with the 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.

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

S. no.	Matrix	Water vapor barrier (transmission rate/permeability)	UV-barrier	Antioxidant activity (%)	Antimicrobial activity against	Coating method	Food item for packaging test	Ref.
1	QCS-TA/OCS	≈14.3 g 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^{-1} s^{-1} 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]

In another approach, taking inspiration from the 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 piece of paper using a layer-by-layer technique followed by oxidative crosslinking using sodium periodate. The modified paper exhibited excellent mechanical properties and water vapor barrier properties. It also 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 nanoparticle-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 a 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. The recent examples of mussel-inspired catechol-based coatings along with a comparison of their characteristic features are given in Table 1.

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 concerns. 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 path-

ways 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^{-1}) in animal models has been associated with gut microbiota disruption, altered bile acid and fatty acid metabolism, inflammation pathway activation, and 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* (Fig. 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^{-1} in food simulants, owing to dopamine-induced cross-linking that halted COF migration.[91] However, research on the toxicity of dopaminated composites is still lacking.

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 m^{-2}).[115] Despite

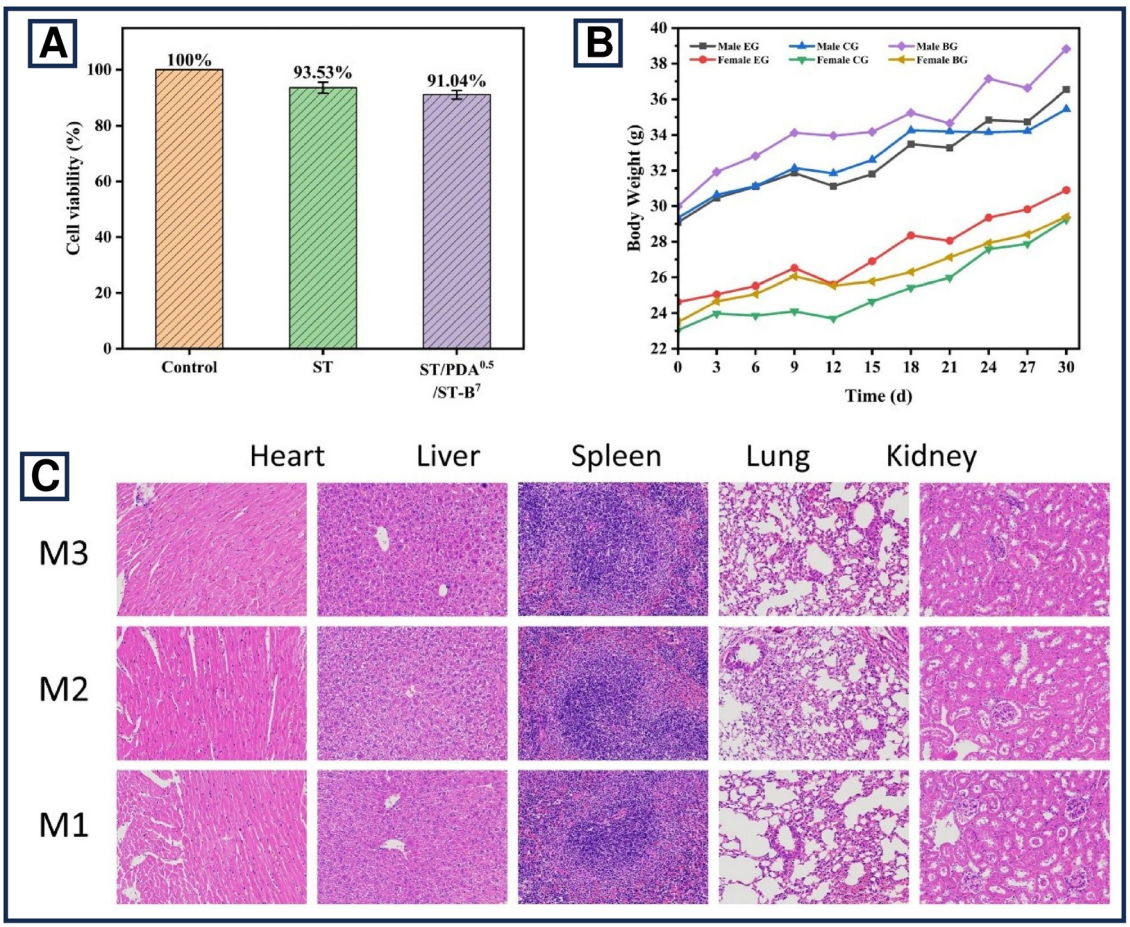

Fig. 12 (A) Effect on the viability of HeLa cells when cultured with neat cornstarch films (ST) and films comprising 93% cornstarch, 3% rosin quaternary ammonium salt-modified cornstarch (ST-B) and 0.5 wt% PDA ($ST/PDA^{0.5}/ST\text{-}B^{7}$) for 3 days. (B) Changes in the body weight of male and female 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 $ST/PDA^{0.5}/ST\text{-}B^{7}$ showing no signs of abnormalities. Adapted with permission from ref. 64. Copyright 2024, Elsevier.

its compliance with permissible non-benzene solvent limits (<5 mg m^{-2}), the degradation products and chemical residues must be carefully evaluated for food contact guidelines.

Edible coatings are subjected to stricter ingestion standards than 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, 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. Bio-based 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-based food packaging films with catechol derivatives, including DA and TA, has gained 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 resulting composite films demonstrated 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 the cross-linking behavior and resulting film properties, necessitates further 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.

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

Data availability

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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