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From Construction to Application: A Comprehensive Review of Protein-Polysaccharide Based Delivery Systems in the Food Industry

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ABSTRACT: Protein-polysaccharide (PRO-POL) complexes or conjugates have shown broad application potential in food research and development because of their favorable digestion-regulating properties and precise targeted delivery capabilities. Nevertheless, a systematic, integrative, and methodologically rigorous review of these systems remains lacking. This review focuses on delivery systems constructed from PRO-POL complexes or conjugates. It first elaborates on the fundamental physicochemical properties and functional characteristics of proteins and polysaccharides, thereby establishing a theoretical foundation for understanding PRO-POL delivery systems. Subsequently, it systematically analyzes the construction strategies of PRO-POL delivery systems, with emphasis on the principles and advantages of mainstream preparation methods, including the Maillard reaction and anti-solvent precipitation. In addition, the facilitating roles of auxiliary technologies, such as ultrasound and microwave treatment, in system formation are discussed, and the effects of key factors, including material ratio, pH, and ionic strength, on delivery efficiency are analyzed. Finally, the practical applications of PRO-POL based delivery systems in the food industry are reviewed, particularly in probiotic encapsulation, dietary fortification, and 3D printing. This review reveals that PRO-POL based delivery system efficiently encapsulates bioactive compounds, thereby enhancing their stability and bioavailability. These findings provide a theoretical basis for further development of the system and facilitate its innovative application within the food industry.

Keywords: Protein-polysaccharide complexes; Bioactive compounds; Preparation methods; Binding mechanisms; Food applications

1. Introduction

In recent years, the food industry has faced persistent bottlenecks in constructing delivery systems for bioactive compounds (BCs), including poor stability, limited controlled release, and low bioavailability. Overcoming the functional limitations of single components and precisely designing carrier structures to achieve the stabilization and targeted delivery of BCs has become a research focus in this field. Among these, PRO-POL delivery systems have attracted widespread attention owing to excellent biocompatibility, environmental friendliness, and tunable structural properties^[1, 2].

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As essential biomolecules, proteins (PRO) execute diverse biological functions stemming from their structural diversity^[1, 3]. Their superior emulsifying properties enable the formation of fine emulsion droplets with narrow size distributions; however, the resulting emulsions exhibit high sensitivity to environmental factors^[4]. Despite their effectiveness as emulsifiers and encapsulation carriers, PRO suffer from practical limitations, notably low solubility and excessive viscosity in many plant-derived variants. In contrast, polysaccharides (POL) provide advantageous characteristics such as biocompatibility, biodegradability, and tunable rheological or gel-forming properties. These features enable adaptation to diverse processing conditions and significantly enhance the functional properties of PRO^[5, 6, 7]. They typically display enhanced water solubility and form denser interfacial layers compared to PRO^[3]. Under specific conditions, electrostatic interactions between charged POL and PRO generate stable complexes or conjugates. These composite structures can be engineered into multifunctional delivery systems including nanoparticles, hydrogels, emulsions, and edible films, facilitating efficient encapsulation and transport of both hydrophilic and hydrophobic BCs. Given their complementary chemical structures and synergistic functional properties, PRO-POL complexes or conjugates serve as versatile platforms for designing advanced delivery systems for BCs.

BCs, such as vitamins^[3, 8], polyphenols^[2, 9], and carotenoids^[10, 11], are poorly water-soluble and sensitive to environmental conditions like light, heat, and oxygen, leading to reduced physicochemical stability. Additionally, their intestinal bioavailability is often marked decrease after oral ingestion. To address these challenges, delivery systems for BCs offer an effective solution. They are designed to embed, encapsulate, or bind BCs, thereby providing protection, targeted delivery, and controlled release^[1]. Given that the development of food delivery systems requires materials with established safety certifications, PRO and POL are widely employed due to their favorable chemical properties. PRO-POL based carrier systems preserve the structural integrity and bioactivity of encapsulated compounds to achieve efficient delivery of BCs. Furthermore, these systems have been proven to effectively mitigate undesirable organoleptic properties of certain BCs, particularly the characteristic astringency of polyphenolic compounds, thereby improving consumer acceptability^[12].

Previous reviews outlined the fundamental composition and formation mechanisms of PRO-POL based delivery systems, along with limited applications in food, chemical, and pharmaceutical industries^[13-15]. These studies provided a theoretical foundation for advancing field. Nevertheless, a systematic discussion regarding the recent advances of PRO-POL based delivery systems for BCs is still absent. Therefore, this review comprehensively summarizes PRO-POL based delivery systems from multiple perspectives: the types and characteristics of wall materials, categories of delivery systems, the construction methods, influencing factors, food applications, current limitations, and future development trends. This review is expected to provide theoretical guidance for the development and application of PRO-POL based delivery systems.

Summary of PRO-POL based delivery systems

PRO-POL based delivery systems exhibit broad applicability in food-related fields, including functional food and dietary supplements, maximizing nutrient bioavailability. Wall materials play a central role in the construction of delivery systems, and their types determine the functional characteristics of final systems. In this section, types and characteristics of wall materials as well as diverse types of PRO-POL delivery systems are fully discussed.

2.1 Types and characteristics of wall materials

2.1.1 PRO

PRO-based delivery systems have demonstrated high encapsulation efficiency for both hydrophilic and hydrophobic nutrients. Natural food PRO, derived from plants, animals, and microorganisms can generate stable delivery systems. Animal-derived PRO, such as bovine serum albumin (BSA) and casein, offer relatively low costs, sustainable advantages, and convenient processing under mild conditions^[16]. However, certain animal PROs, such as gelatin, exhibit relatively poor gelation properties, high pH sensitivity, and a considerable environmental burden from large-scale production^[17]. Plant PRO, such as zein and soy protein isolate (SPI), possess stronger hydrophobicity, lower toxicity risk, superior sustainability, reduced allergenicity, and greater cost-effectiveness compared to animal PRO^[3, 18]. These advantages have prompted the gradual transformation of plant PRO as promising substitutes for animal PRO. In contrast, microbial PRO is less commonly used in delivery systems due to structural complexity, limited intrinsic targeting capability, and the immaturity of relevant processing technologies. Nevertheless, recent studies have explored *Spirulina* protein isolate^[19] and *Haematococcus pluvialis* protein (HPP)^[20] as potential raw materials. Owing to the excellent biocompatibility and favorable structural properties, PRO have been extensively studied and widely utilized in BCs delivery. The details are shown in Table 1.

Table 1 Types and characteristics of PRO in delivery systems

PRO types	pI	Characteristics	Typical Applications	References
Animal PROs				
Ovalbumin (OVA)	4.5	Basic protein, lipid-soluble, heat-induced deformation.	Bakery ingredient, meat binder, foaming dessert.	[2, 17, 21]
Lactoferrin (LF)	6.0 (human) 8.0 (bovine)	Alkaline protein, stable in the gastrointestinal tract, antimicrobial.	Nutritional fortifier, natural preservative, infant formula.	[22]
BSA	4.7	Water-soluble, heat sensitive.	Emulsion model, foam stabilizer.	[23, 24]
Whey protein isolate (WPI)	5.2	Thermal deformation (>62°C), aggregating and precipitating under acidic conditions, ordered globular structure	High-PRO beverages, flavor encapsulant, emulsifier and gelling agent.	[2, 25, 26]
β -lactoglobulin (β -LG)	5.0	Water (acidic pH)-soluble, thermal denaturation temperature of 70°C at neutral pH, fat-binding PRO.	Clear beverage ingredient, foaming agent, yogurt gel enhancer.	[27, 28]
Gelatin	A 7.0-9.0 B 4.7-5.2	Water-soluble (>40°C), forming gels (<37°C), thermal stability.	Confectionery, yogurt thickener, meat gelling, beverage clarifier.	[29, 21, 30]
Casein	4.6-4.8	Soluble in neutral and alkaline solutions (sodium caseinate), thermal stability.	Cheese base, bakery fortifier, creamer emulsifier, meat fat stabilizer.	[16, 31]
Lysozyme	10.7	Surface hydrophobicity, thermo-gel	Cheese/wine preservative, fresh	[32]

		properties, antimicrobial.	produce antimicrobial coating.	
Vegetable PROs				
Zein	6.2	Ethanol-soluble, thermal stability, poor physical stability.	Gum base, gluten-free dough network.	[18, 2, 21]
Soy protein isolate (SPI)	4.0-5.0	Alkaline-soluble, poor <u>emulsification</u> , sensitivity to environment, mask undesirable flavors.	Plant-based meat/dairy, nutrition bars.	[21, 33]
Peanut separated protein (PSP)	4.3-4.5	Poor solubility and emulsification.	Nut-flavored beverages, cereal bars, bakery fortifier.	[33, 34]
Pea protein	4.5-5.2	Soluble at acidic pH, unpleasant soya odor, sustainability, hypoallergenicity.	Vegan meat/dairy, sports protein.	[33, 35, 36]
Microbial PROs				
Yeast protein	4.0-5.0	Good physiological functions and processing characteristics, high digestion efficiency.	Flavor enhancer, cheese base, meat texture modifier.	[37]
HPP	4.2	Antioxidant	Antioxidant, beverage stabilizer.	[20]
<u>Phycocyanin</u>	3.4	Water-soluble, antioxidant, anti-inflammatory, easy to degrade, poor colloidal and color stability	Natural blue colorant, antioxidant.	[38]
<i>Spirulina</i> protein isolate	3.0	Solubility at alkaline pH.	Meal replacement, colorant.	[19]

2.1.2 POL

POL possesses numerous sources, including animal tissues, plant cell walls, and microbial secretions, exhibiting significant variations in molecular weights, structural configurations, physicochemical properties, and functional characteristics (Table 2). The primary chains of POL contain various carboxyl, hydroxyl, amino, and aldehyde groups, that can form delivery systems via physical interaction or chemical cross-linking^[39]. POL is able to modify and extend the functional characteristics of PRO, thereby improving the stability of PRO delivery systems and modulating PRO-POL interactions^[23].

Animal POL exhibits excellent cytocompatibility, rendering it a common component for delivery systems. Plant POL features low processing costs and aligns with green development principles, effectively addressing the challenges posed by animal POL. Moreover, plant POL possesses strong intermolecular interactions and inherently stable structures that resist deformation under changing environmental conditions, making it well suited as stabilizers in food emulsions^[40]. Nevertheless, limitations such as low activity of plant POL affect production efficiency and scalability^[41]. Compared with animal and plant POL, microbial POL offers higher production efficiency, stronger structural stability, lower environmental impact and cost^[42].

Table 2 Types and characteristics of POL in delivery systems

POL types	Molecular weights	Characteristics	Typical Applications	References
Animal POLs				
Hyaluronic acid (HA)	5-1800 kDa	Ethanol-soluble, hydrophilic.	Health supplements.	[9, 43]
Chitosan (CS)	300-1000 kDa	Soluble in dilute acid solutions below pH 6.3, antibacterial, poor thermal stability.	Antimicrobial packaging, beverage clarifier.	[44]
Chitin	400-1600 kDa	Insoluble in water, pH-sensitive.	Thickener.	[13, 45]

Plant POLs				
Pectin	50-150 kDa	Hydrophilic, pH-sensitive, easy to modify.	Gelling agent, acid dairy stabilizer, fat replacer.	[44, 46]
Gum Arabic (GA)	0.03-1000 kDa	Good solubility and emulsifying properties, heat-resistant.	Confectionery anti-crystallizer, beverage flavor emulsifier.	[47, 44]
Dextran	100-10000 kDa	Neutral POL, good solubility and stability.	Ice cream/jam thickener, anti-crystallizer.	[34]
Starch	Amylose: 80-1000 kDa Amylopectin: 10-100 MDa	Low mechanical strength, high hydrophilicity, amylose is semi-crystalline and soluble in hot water, amylopectin is very crystalline and does not dissolve in water.	Texturizer, fat replacer, biodegradable packaging, thickener.	[44, 46, 48]
Cellulose	10-1000 kDa	Water-insoluble, stable, modified or combined with others to serve as a carrier.	Cheese anti-caking, fat replacer.	[44, 46]
Soybean polysaccharide	50-1000 kDa	Water-soluble, low-viscosity, heat-stable.	Acid beverage stabilizer.	[23, 47]
Agar	80-240 kDa	Water-soluble, forming a gel upon cooling.	Canned meat gel, bakery glaze, confectionery stabilizer.	[47, 49]
Konjac glucomannan (KGM)	200-2000 kDa	Water-soluble, stable in acidic conditions, strong water-absorbing capacity.	Low-calorie noodles, fat replacer.	[50, 51]
Microbial POLs				
SA	100-600 kDa	Water-soluble, hydrophilic, acid-resistant, weak stability, heat-instability.	Ice cream stabilizer, thickener.	[52, 46]
Xanthan gum (XG)	2-20 MDa	Water-soluble, enzymatic degradation resistance, agglomerating during slow dissolution in cold water.	Thickener, stabilizer, texture modifier.	[46, 53]
Pullulan	40-600 kDa	Water-soluble, heat-resistant.	Candy coating, food binder.	[26, 54]
Gellan gum (GG)	25-288 kDa	pH stable, low charge density.	Heat-resistant jelly, stabilizer, fluid gel beverage.	[46, 55]

2.2 Types of PRO-POL based delivery systems

Owing to the complementary physicochemical properties, PRO and POL can form structurally diverse and functionally distinctive complexes or conjugates. Depending on their structural organization and preparation methods, PRO-POL based delivery systems can be categorized into several major types, including microcapsules, nanoparticles, gels, films, and emulsions, etc. These systems differ in encapsulation efficiency, physicochemical stability, release behavior, and applicability to various BCs. Here, representative delivery systems are displayed in Fig. 1 and Table 3.

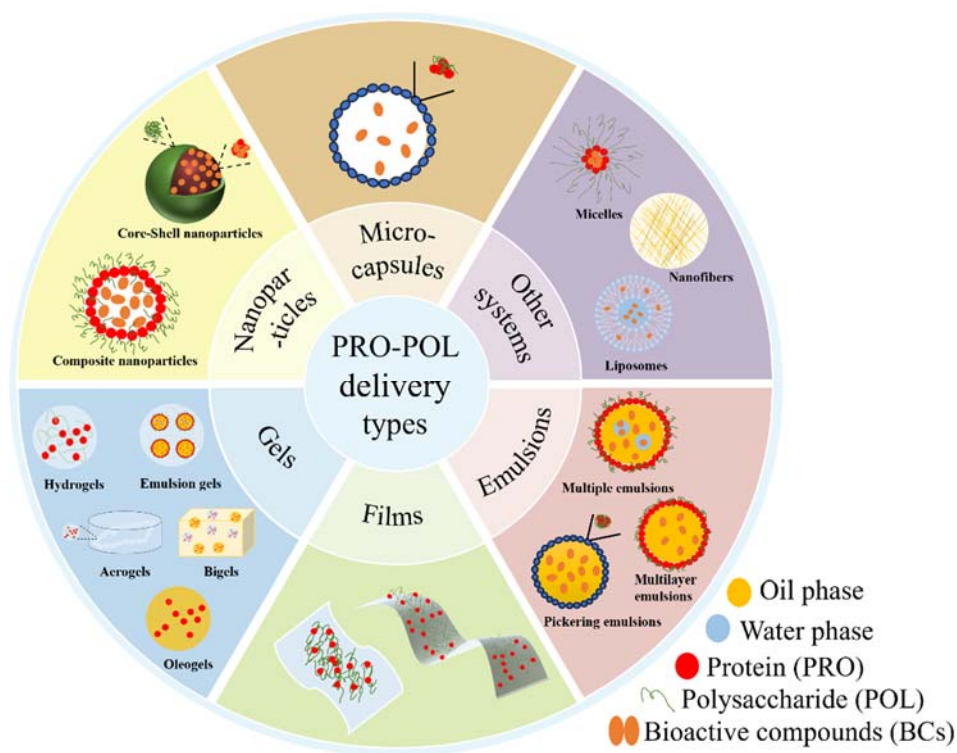


Fig. 1. Common types of PRO-POL based delivery systems.

Table 3 Summary of PRO-POL based delivery systems

<i>PROs</i>	<i>POLs</i>	PRO-POL based systems	Encapsulated ingredients	Functions	References
Zein	XG	Zein-XG-VE microcapsule	VE	High encapsulation efficiency; excellent thermal stability; and sustained release.	[8]
Zein	Pectin	Zein-pectin nanoparticle	Tanshinone	Enhancement of Tanshinone solubility.	[56]
HPP	Galactose (GAL)	HPP-GAL nanoparticles	Curcumin (Cur)	Improvement of encapsulation efficiency and loading capacity; good antioxidant activity in vitro and Cur release.	[57]
WPI	Pullulan	WPI-Pullulan β -Carotene emulsions	β -carotene	Extended β -carotene's half-life.	[26]
WPI	Pectin	WPI-Pectin-VD ₃ nano-emulsion	VD ₃	Fortified sunflower oil by enhancing bioavailability and shelf life.	[58]
SPI	Soybean oligosaccharide	SPI-soybean oligosaccharide hydrogels	Zeaxanthin	Higher photochemical stability and bioavailability of zeaxanthin; slower chemical degradation.	[11]
SPI	Corn starch	SPI-corn starch- β -carotene	β -carotene	Showing flexible structure, high printing accuracy, and strong mechanical properties.	[59]
C-phyco cyanin	Pectin	C-phyco cyanin-pectin-polyphenol based hydrogels	<i>L. plantarum</i>	Enhancement in the viability of <i>L. plantarum</i> under gastrointestinal conditions.	[38]
Sodium	CS,	SC-CS-pectin	Lutein	Improvement of lutein retention and	[60]

caseinate (SC)	pectin	multilayer emulsion			stability.
Yeast protein	CS	Yeast protein-CS composite emulsions	Monascus pigments (MP)	High encapsulation of MP; improvement of color retention and storage stability. [37]	
Soy whey protein (SWP)	SA	SWP-SA emulsion	Soybean isoflavones	Increasing the bioavailability of soy isoflavones. [61]	
OVA	Fucoidan (FUC)	OVA-FUC binary complexes O/W emulsions	Egg yolk lecithin	Enhancement of delivery efficiency, and stable behavior in vitro digestion. [62]	
HPP	CS	HPP-CS Pickering emulsion	Astaxanthin	Reduced sodium intake; and strong antioxidant capacity. [20]	
Myofibrillar protein	CS	Myofibrillar protein-CS emulsion	Astaxanthin	Preventing the degradation of astaxanthin during storage. [63]	
Glycated casein (UGCA)	Carboxy methyl cellulose (CMC)	ACNs-UGCA-CMC ternary complexes	Anthocyanins (ACNs)	Strengthening the stability; and controlled release of ACNs. [64]	
WPI	Maltodextrin, inulin	WPI- Maltodextrin-inulin complexes	<i>Saccharomyces pastorianus</i>	Enhancement of cell protection during fixation and microbial stability. [65]	

2.2.1 Microcapsules

Microcapsules represent an innovative technological approach that encapsulates BCs (minute particles of gases, liquids, or solids) within biopolymeric wall materials via specific physical, chemical, or mechanical techniques. This approach yields particles ranging from the nanometer to millimeter scale in diameter, exhibiting diverse morphologies and internal structures^[66]. Microcapsules are designed to protect and preserve the encapsulated substances from environmental factors, and enable controlled release under gastrointestinal or physiological conditions^[67]. Moreover, they can protect desirable flavors and aromas or mask unpleasant odors. Selecting appropriate coating materials is essential for achieving controlled release, maintaining BCs stability, and improving microcapsule quality. Numerous techniques have been developed to prepare BC-loaded microcapsules such as coacervation, emulsification, polymerization, spray-drying, freeze-drying, and melt-extrusion^[68].

In laboratory practice, microcapsules are typically prepared as single-layer systems. However, BCs in single-layer microcapsules are prone to degradation under harsh storage conditions, compromising release profiles and limiting payload capacity^[69]. Consequently, a range of double-layer microcapsules has been developed to overcome the aforementioned limitations. WPI-SA double-layer microcapsules produced by spray drying enhanced protection and storage stability for encapsulated flaxseed oil^[69]. Another approach employed gelatin and gum arabic as the primary wall materials with a secondary shell containing nano-SiO₂

to encapsulate anthocyanin, further improving encapsulation efficiency, thermal resilience and storage stability^[70]. Thus, double-layer microcapsules exhibit higher structural integrity, antioxidative stability and heat resistance than single-layer systems, making them promising candidates for nutrient preservation in infant formula during storage. Introducing an additional outer layer enhances barrier properties, reduces oxygen and moisture permeation, and regulates release behavior. Yet, high costs and complicated processes limit further commercial applications. A primary limitation of microcapsules is that high encapsulation efficiency does not necessarily guarantee prolonged storage stability since some degradation remains unavoidable, and encapsulation efficiency decreases with increasing payload capacity.

2.2.2 Nanoparticles

Nanoparticles, as a major type of nanoencapsulation, are generally in the nanoscale range, which minimizes aggregation and precipitation, while achieving controlled or targeted release^[71, 72]. Hydrophobic interactions between PRO and POL enhance nanoparticles stability under varying conditions, improve thermal resistance, and enable controlled release of the encapsulated BCs^[47, 73]. Among the numerous strategies for constructing PRO-POL nanoparticles, heat treatment and pH regulation are the most commonly employed. The formation of PRO-POL nanoparticles is highly sensitive to key thermal processing parameters and the inherent properties of the solution^[72]. Cur nanoparticles were prepared using LF-HA-EGCG exhibited improved thermal, photo, and storage stability, bioavailability of Cur, as well as excellent redispersibility after dehydration^[9]. In addition to serving as delivery carriers, nanoparticles also play a stabilizing role in emulsion and foam systems, thereby enhancing the performance of these systems.

Core-shell nanoparticles, a representative type of nanoparticles, are typically fabricated by first forming a PRO core, followed by coating with a POL shell. The PRO core furnishes a favorable microenvironment that protects encapsulated BCs, while the hydrophilic POL shell enhances particle stability through steric hindrance and electrostatic repulsion^[74]. Combining biopolymers with different polarities enables the construction of core-shell structures with hydrophobic-hydrophilic or hydrophilic-hydrophobic configurations^[74]. Among these, nanoparticles with a hydrophobic core and a hydrophilic shell are particularly effective for embedding non-polar BCs in aqueous environments^[75]. Chen et al.^[75] established a core-shell nanoparticles for co-encapsulating coenzyme Q10 and piperine through the combined use of hydrophilic *k*-Car and hydrophobic zein. This strategy improved the stability of encapsulated compounds and validated the efficacy of the resulting nanoparticles. However, the performance of nanoparticles varies considerably, as key parameters such as BCs-loading capacity and stability strongly depend on the particular structural configurations and manufacturing conditions.

2.2.3 Gels

PRO-POL based delivery systems generate composite gel structures, including hydrogels, aerogels, emulsion gels, oleogels and bigels, with multi-scale organization, gradient properties, and enhanced nutritional value^[76].

Hydrogels are three-dimensional network structures formed by crosslinked polymer chains through physical, ionic, or covalent interactions. They exhibit large surface area, high water absorption capacity, porous structure, excellent shape adaptability, outstanding biocompatibility, as well as mechanical stability^[77, 19]. Hydrogels can be classified into natural, synthetic, and semi-synthetic hydrogels depending on the polymer source. In food processing, hydrogels are primarily used to modulate fat content, alter texture and rheological properties, and encapsulate key ingredients^[78]. A previous study reported that a composite hydrogel prepared by β -LG and propylene glycol alginate was used to encapsulate *L. rhamnosus* and Cur, which enhanced probiotic viability, delayed Cur release and improved its chemical stability^[79]. Bu et al.^[80] fabricated a hydrogel using hawthorn pectin and *Tenebrio molitor* protein. This hydrogel exhibited a dense, homogeneous structure with fine pores and enhanced the stability of encapsulated Cur. The above studies demonstrated that hydrogels with stable network structures can encapsulate environment-sensitive BCs and probiotics, thereby enhancing physicochemical stability and survival rate. Meanwhile, the network diameter of hydrogel can be modulated in response to environmental factors, enabling controlled release of encapsulated substances under specific conditions^[23]. Owing to their limited tensile strength and the difficulty of stably dispersing of lipophilic BCs in aqueous matrices, hydrogels are not optimal carriers for such compounds^[81].

In recent years, aerogels have attracted considerable interest due to their broad application potential, including BCs delivery systems, nutritional fortifiers, flavor enhancers, and salt, sugar, or fat alternatives. Aerogels are ultra-lightweight materials consisting of microporous networks in which gas constitutes the dispersed phase^[82]. Depending on the drying technique, dried gels are classified into aerogels, cryogels, and xerogels^[83]. Xerogels are characterized by a dense structure, whereas cryogels demonstrate enlarged porosity and reduced specific surface area. Aerogels development targets the fabrication of advanced materials addressing inherent drawbacks of conventional hydrogels, including excessive water retention, handling difficulties during storage/processing, and limited controllability of pore architecture^[82, 84]. Recently, Mekale et al.^[85] fabricated an aerogel that exhibited high stability and effectively prevented vitamin degradation from pectin and fava bean protein concentrate. Owing to the unique advantages, aerogels hold considerable promise for future food applications.

Emulsion gels feature a unique three-dimensional network structure that combines the advantages of both emulsions and gels. Droplets immobilized in the gel network enable the simultaneous loading of hydrophilic and lipophilic BCs and prevent droplet flocculation or coalescence. They offer considerable potential for improving thermo-dynamic and chemical stability, biocompatibility, as well as texture modification^[86]. PRO-POL based emulsion gels as matrices for encapsulating BCs and enhancing gastrointestinal absorption have attracted increasing attention in the food industry. For example, the emulsion gels used as solid fat replacers were produced with *k*-Car, PPI, and sunflower seed oil^[87]. In addition, due to adjustable composition, viscoelasticity, favorable rheological properties, and recoverability, emulsion gels can be used to make edible inks for 3D printing^[88, 89]. 3D printing ink rich in β -carotene was

prepared from SPI, corn starch, and Ora-pro-nobis mucilage, showing flexible structure, high printing accuracy, and strong mechanical properties^[59]. Yet, the parameter window for emulsion gels is narrow, and an excessively dense network can impede digestion.

Oleogels are semi-solid systems composed of three-dimensional networks formed via molecular self-assembly and intermolecular interactions^[81]. In contrast to hydrogels, oleogels serve as effective delivery vehicles for lipophilic BCs and present a viable strategy for replacing solid fats in food products. Zein-XG oleogels, when used as fat substitutes, exhibited sponge-like porous bridged network structure, as well as underwent minimal degradation during prolonged heating in medium^[90]. Oleogels offer favorable mechanical properties to protect encapsulated lipophilic BCs^[55, 81]. Based on the lipophilicity of oleogelators, preparation methods can be classified into the direct method for hydrophobic oleogelators and the indirect method for hydrophilic oleogelators. Due to the limited capacity to deliver hydrophilic BCs and potential safety concerns, oleogels currently have fewer applications in the food industry compared to other gels.

Bigels overcome the inherent limitations of hydrogels and oleogels, which are confined to a single aqueous or lipid phase. This unique structure synergistically integrates the advantages of both phases, enabling the co-delivering of hydrophilic and lipophilic BCs^[83]. Depending on the formulation and phase ratio, bigels can be classified as oleogel-in-hydrogel, hydrogel-in-oleogel, bicontinuous double gels, or more complex multi-domain structures. Gelation of both water and oil phases immobilizes droplets within the three-dimensional network, preventing coalescence and flocculation, thereby markedly improving stability. Moreover, increasing the hydrogel fraction enhances hydrogen-bond density, which raises rigidity and viscosity, and further boosts its potential as delivery vehicles^[83]. Since bigels are still at an early development stage, their application in food systems remains to be fully explored^[81].

2.2.4 Films

PRO-POL film can be classified into barrier films, active packaging films, edible/biodegradable films, and smart films. These films exhibit superior physical properties, including improved water vapor and oxygen barrier performance as well as greater mechanical strength, compared to single-component films^[91]. Active packaging films comprise three primary categories: antioxidant films, antimicrobial films, and functional delivery carriers. Incorporating natural polyphenols into these films effectively inhibits oxidative processes^[92]. The addition of natural antimicrobial agents imparts antibacterial functionality to the films^[93-95]. Furthermore, these films serve as carriers for BCs, enabling both encapsulation and controlled release^[96]. Edible and biodegradable films offer sustainable alternatives for coating and preserving fresh produce, providing a viable route to replacing conventional plastic packaging^[97]. Recent advances include the development of multifunctional smart packaging films based on cross-linked corn protein matrices, which combine pH responsiveness with antimicrobial and antioxidant activities^[98]. These intelligent materials enable real-time monitoring of meat freshness via integrated mobile applications, demonstrating the convergence of active packaging and information technology. Notably, these functional classifications remain

flexible in practice, as most films simultaneously exhibit multiple functionalities, thus providing a theoretical basis for designing integrated multifunctional film systems.

2.2.5 Emulsions

PRO-POL based emulsions serve as versatile platforms for encapsulation and delivery of BCs. These emulsion systems, encompassing Pickering emulsions, multiple emulsions, and multilayer emulsions, leverage synergistic interactions to enhance encapsulation efficiency and environmental responsiveness.

Pickering emulsions offer high stability, low toxicity, anti-agglomeration properties, biodegradability, as well as higher environmental tolerance than conventional emulsions^[99, 100]. Pickering stabilization arises from the accumulation of dispersed particles at the oil-water interface, forming a mechanical barrier that protects droplets against coalescence^[100]. Generally, the emulsions stabilized only by PRO are sensitive to temperature, pH, salt, and environmental stresses, which result in coalescence, flocculation, creaming, and phase separation^[64]. Incorporating POL can enhance the stability of PRO-based emulsions during storage, as well as improve the absorption and transport of hydrophobic compounds^[101]. WPI-dextran Pickering emulsion confirmed that the binding of protein and polysaccharide significantly improved the bioavailability and cellular uptake of Cur^[102]. The finding highlights the potential of Pickering emulsions as a promising delivery platform to enhance the bioavailability and stability of BCs.

Multiple emulsions (e.g., W/O/W and O/W/O) represent a distinct class of emulsions formed by integrating W/O and O/W systems, thereby combining the advantages of both. These emulsions are characterized by high encapsulation efficiency, robust physical stability, and controlled release properties. Moreover, they not only allow for the simultaneous encapsulation and co-delivery of nutrients, BCs, and flavor substances for controlled release^[103] but also establish W/O/W systems to reduce fat content^[23]. Casein-SA- β -cyclodextrin multiple emulsions (W/O/W) were successfully prepared for co-encapsulation iron and Cur, which enhanced emulsion interfacial stability and bioaccessibility^[104]. The preparation involves a two-step emulsification process transitioning from a W/O pre-emulsion to a W₁/O/W₂ final emulsion, thereby generating diverse structural hierarchies and necessitating precise control of multiple processing parameters. Furthermore, these emulsions suffer from structural complexity and are primarily suited for fortifying liquid foods, which restricts their use in solid matrices.

Multilayer emulsions often exhibit excellent physical stability under various environmental stresses, which is attributed to the electrostatic interactions between oil droplets through multilayer interfacial films^[105, 106]. Generally, these interfacial films consist primarily of emulsifiers (PROs) and charged biopolymers (POLs)^[106]. Multilayer emulsions are constructed through the fine regulation of interfacial architecture, trading structural complexity for enhanced stability. Their preparation demands precise control over layer-by-layer deposition while preventing bridging flocculation caused by an insufficient outer POL concentration^[103, 107]. Shi et al.^[60] successfully prepared a CS-SC-pectin multilayer emulsion for lutein encapsulation, which significantly improved the retention and stability of lutein. Consequently, multilayer emulsions have emerged as a compelling platform for BCs delivery in food systems. Based on existing

literature, a comparative analysis of the advantages and limitations of each delivery system type is provided in Table 4.

Table 4 Comparative summary of each delivery system

Types	Merits	Drawbacks	References
Microcapsules	High encapsulation efficiency and stability, controlled release, protecting desirable taste and aroma, or masking unpleasant odors.	Insufficient storage stability, relatively low operational controllability, mainly embedding hydrophobic BCs.	[67, 108]
Nanoparticles	High encapsulation efficiency and environmental stability, controlled release.	Hydrophobic BCs embedding, performance governed by structural configurations and fabrication conditions.	[9]
Hydrogels	Large surface area, high water absorption capacity, porous structure, excellent shape adaptability, outstanding biocompatibility, and mechanical stability.	Mainly embedding hydrophilic BCs, limited tensile strength, excessive water retention, handling difficulties during storage/processing.	[21, 103, 108]
Aerogels	High porosity, large specific surface area, low solid density.	Insufficient structural stability and mechanical properties, complex preparation process.	[82, 84]
Emulsions	Coordinated biphasic loading of hydrophilic and hydrophobic BCs, enhanced physicochemical stability, and biocompatibility, texture modification	Narrow parameter window, excessively dense networks may hinder digestion	[86]
Oleogels	Optimal mechanical properties, high gel strength and stability, masking of undesirable flavors of nutraceuticals.	Mainly embedding hydrophobic substances, limited applications.	[81, 55, 14]
Bigels	Coordinated biphasic loading of hydrophilic and hydrophobic BCs, synergistically combining the advantages of hydrogels and oleogels.	Limited applications by preparation.	[81, 83].
Films	Greater mechanical strength, strong barrier properties, antibacterial and antioxidant, extend shelf life	Fabrication predominantly via electrospinning, precise operation required.	[94, 91, 92, 95]
Pickering emulsions	High stability, low toxicity, anti-agglomeration, biodegradability, and higher environmental tolerance, mechanical barrier stability, delivery of both hydrophobic and hydrophilic BCs.	Difficulty in achieving balance during preparation. challenges in reaching target droplet sizes, limiting industrial yield, and unimproved biocompatibility.	[99, 14, 100, 108]
Multiple emulsions	High encapsulation efficiency, robust physical stability, and controlled release; coordinated biphasic loading of BC.	High cost, poor storage stability, complex preparation, and limited to liquid food fortification.	[14, 108]
Multilayer emulsions	Positive physical stability and bioaccessibility, optimal storage stability, fish oil bioaccessibility, controlled release.	Complex preparation requiring precise control of processing parameters.	[103, 109]

2.2.6 Other delivery systems

In addition to the aforementioned common PRO-POL based delivery systems, several other emerging systems, like micelles, nanofibers, and liposomes, can also be utilized for the encapsulation and delivery of BCs.

Micelles are nanoscale aggregates that form spontaneously from amphiphilic polymers in aqueous solution when the critical micelle concentration is reached; in these structures, PRO and POL serve as the hydrophobic and hydrophilic moieties, respectively^[110]. PRO-POL nanofibers fabricated by electrospinning, with high porosity, large specific surface area, and unique physicochemical properties, can also be utilized

for BCs delivery. Zhao et al.^[111] prepared uniform nanofibers from tremella polysaccharide and SPI via electrospinning, which were expected to be applied in food packaging films, representing an emerging trend in food packaging nanomaterials. Liposomes incorporating PRO and POL have garnered interest due to their ability to encapsulate both hydrophilic and hydrophobic BCs. For instance, a bio-nanocomposite film composed of gelatin, CS, and tobacco extract-loaded nanoliposomes extended the shelf life of perishable foods^[96]. The formation pathways and underlying mechanisms of PRO-POL based delivery systems vary considerably depending on the characteristics of each system. These mechanisms are analyzed systematically in the following section.

3. Preparation methods and influencing factors of PRO-POL based delivery systems

The combination of PRO and POL occurs via chemical covalent bonding or non-covalent physical interactions. Generally, covalent interactions yield PRO-POL conjugates (PPCOs), whereas non-covalent interactions produce PRO-POL complexes (PPCs). Both PPCOs and PPCs serve as effective delivery carriers, enhancing the stability, bioavailability, and overall efficacy of BCs. Common preparation methods for PPCOs and PPCs are summarized in this section and illustrated in Fig. 2.

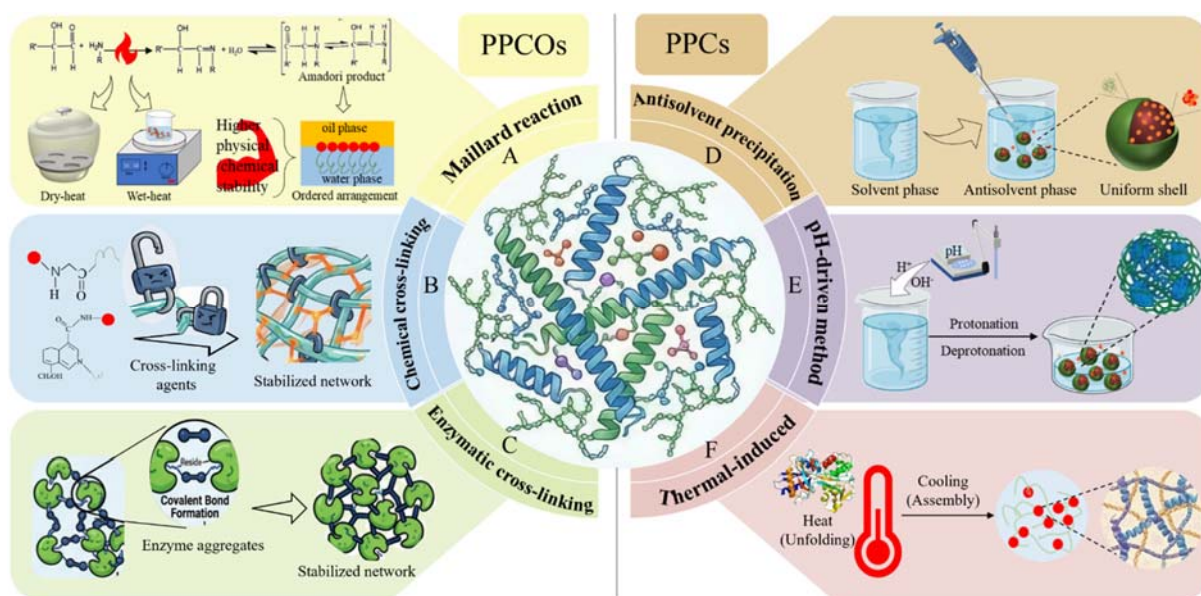


Fig. 2. Common binding methods of PPCOs and PPCs.

3.1 Preparation methods of PPCOs

The covalent conjugation between POL and PRO provides an effective platform for constructing PPCOs. These conjugates enhance steric stabilization, structural robustness, and emulsifying capacity of the resulting systems^[112]. In addition, PPCOs maintain effective emulsion stability for several weeks even after substantial dilution, as well as markedly improve resistance to environmental stresses^[113]. Covalent interactions are highly specific, establishing permanent binding and irreversible interaction between PRO and POL^[23]. Compared with non-covalent complexes, covalent conjugates are insensitive to changes in pH and ionic strength^[113].

PPCOs can be constructed via several techniques, primarily the Maillard reaction, chemical cross-linking, and enzymatic cross-linking^[23, 112]. Each method exhibits distinct advantages and limitations, making it imperative to comprehensively evaluate both the intrinsic physicochemical properties of the encapsulated compounds and the specific environmental conditions they will encounter. Such systematic assessment enables the rational selection of optimal preparation methods to ensure maximal efficacy for BCs delivery.

3.1.1 Maillard reaction

The Maillard reaction, a non-enzymatic browning process typically initiated by thermal treatments like dry- or wet-heating, is a primary method for preparing PPCOs due to its mild conditions and suitability for food-grade applications(Fig. 2A)^[114]. In this reaction, free amino groups (e.g., from lysine residues) of proteins react with the reducing-end aldehyde group (-CHO) of polysaccharides to form Schiff bases, which subsequently rearrange into stable structures^[115, 1, 116]. This reaction yields PPCOs with high thermal stability, improved water solubility, antibacterial capacity, and antioxidant activity^[115, 117]. However, the Maillard reaction poses inherent risks, particularly during its intermediate and advanced stages. At this stages, melanoidins and advanced glycation end products are generated, which are closely linked to various chronic diseases. Meanwhile, abnormal cross-linking and thermal aggregation between protein chains may be induced, resulting in phase separation and precipitation of the complex. Furthermore, regardless of the heating mode, thermosensitive compounds may be degraded or deactivated by the high thermal stress required for glycosylation^[118]. Thus, recent research has focused on the early-stage condensation between the reducing-end carbonyl groups of POLs and the amino groups of PROs, which proceeds with the release of water. By optimizing reaction parameters such as time, pH, water activity, and temperature, the reaction process can be effectively controlled to prevent the formation of anti-nutritional and toxic compounds^[117].

PPCOs prepared via the Maillard reaction enhance emulsion stability by restructuring oil-water interfaces and can encapsulate hydrophobic BCs^[101, 119]. When stabilizing algal oil emulsions, PPI-inulin conjugates yielded exceptional physical and oxidative resilience. Jiang et al.^[120] demonstrated that the protective effect stemmed from profound steric repulsion exerted by hydrophilic inulin chains, coupled with augmented antioxidant capacity at PRO-rich boundaries. Zhang et al.^[101] successfully expanded such robust interfacial engineering toward Pickering systems. The research showed PPI conjugated with okra flower polysaccharides constructed a highly robust interfacial layer, essentially fortifying lipid-protein affinity. Consequently, when utilizing this sterically hindered matrix to encapsulate apigenin, it acted as a formidable barrier, enabling precisely controlled release throughout the gastrointestinal digestion. To achieve the desired properties and optimal encapsulation efficiency, the Maillard reaction must be carefully controlled to avoid undesirable by-products that compromise conjugate purity and yield unsatisfactory results^[121]. Meanwhile, it should be noted that excessive masking of lysine residues by POL can significantly diminish the associated functionality of the PRO. Common strategies to mitigate this issue include incorporating

high-affinity polyphenols, selecting modified low-molecular-weight or low-esterified polysaccharides, and applying physically assisted techniques to preserve bioactive lysine residues.

3.1.2 Chemical cross-linking

The covalent bonds formed by chemical cross-linking significantly enhance the mechanical strength, environmental tolerance, and biological stability of the PPCOs (Fig. 2B). Such cross-linking typically involves the addition of cross-linking agents like GA, genipin, EDC and DMTMM. Among these, GA is prohibited in the food industry owing to its inherent toxicity. Genipin is a water-soluble and plant-based cross-linking agent with antioxidant and anticancer properties^[122], which covalently binds primary amino groups of PRO and POL via nucleophilic substitution reaction. Razi et al.^[123] developed genipin-crosslinked caseinate-CS nanocomplexes for Cur delivery. The cross-linking process inherently depleted primary amines, driving notable reduction in zeta potential (from +32.6 to +24.7 mV). Consequently, covalent modification shielded the nanocarriers against structural disruption caused by pH and ionic strength, thereby markedly enhancing in vitro anticancer activity and sustained stability. These findings demonstrated the potential of genipin-mediated cross-linking to improve delivery efficiency of BCs. Cross-linking with EDC yields PPCOs exhibiting superior mechanical stability and diminished cytotoxicity than GA-based cross-linking^[124]. In addition, EDC and DMTMM mediate the formation of stable amide bonds between primary amines ($-NH_2$) and carboxyl groups ($-COOH$), integrating into the network structure. These compounds are converted into small molecular byproducts that can be removed through dialysis or other purification methods^[122].

However, removing residual cross-linking agents requires additional post-processing steps, which complicates the procedure and substantially prolongs the preparation time. Furthermore, excessive cross-linking can produce an overly dense network structure that impedes the effective release of BCs.

3.1.3 Enzymatic cross-linking

Enzymatic cross-linking, refers to enzyme-catalyzed covalent bond formation between reactive groups in PRO and POL, thereby generating stable biopolymer networks or conjugates (Fig. 2C). It offers noticeable advantages, including short reaction times, mild processing conditions, high substrate specificity, and minimal by-product formation^[23, 77]. These features render enzymatic cross-linking particularly appealing for food industry applications where both food safety and environmental sustainability are important. Various enzymes such as polyphenol oxidase, peroxidase, and transglutaminase (TG) have been employed to promote covalent cross-linking reactions between PRO and POL, either at fluid interfaces or in bulk phases^[23]. These enzymes generate a reactive intermediate that spontaneously polymerizes with other groups to form covalent crosslinks rather than directly linking PRO^[125]. Enzymes cleave specific peptide bonds in PRO chains, enabling exposed amino acid groups to form covalent bonds with phenolic components of POL, thereby stabilizing the gel network^[1, 126]. Among these, laccase is the most environmentally friendly, as it generates only water as a by-product. Laccase and peroxidase can oxidize

various phenolic and aniline compounds via a single-electron removal mechanism, generating free radicals through the reduction of molecular oxygen^[125]. TG links a lysine ϵ -amino group (acyl acceptor) to a glutamine γ -carboxamide group (acyl donor) in proteins^[125, 127]. Notably, TG has also been employed to facilitate cross-linking between PRO and POL bearing amino groups. In gliadin-CS co-precipitated nanoparticles, TG significantly enhanced the colloidal stability and interfacial activity by catalyzing the irreversible covalent bonding between amino groups of CS and the glutamine residues of gliadin via acyl transfer reactions^[128]. Generally, enzymatic cross-linking not only strengthens PRO-POL interactions but also increases the viscosity of biopolymer solutions, potentially leading to gel formation and improved functional properties^[23, 77]. A key limitation of enzyme-mediated cross-linking is its pronounced dependence on enzyme dosage, reaction time, and physical state of the substrate protein. Minor fluctuations in any of these factors may compromise the intended cross-linking efficacy.

3.2 Preparation methods of PPCs

PPCs are primarily formed through non-covalent interactions, including electrostatic interactions, hydrophobic interactions, hydrogen bonds, and van der Waals forces^[115, 129]. Although these physical associations yield inferior mechanical properties compared with covalent linkages, they offer notable advantages for food applications owing to their spontaneous formation, procedural simplicity, and low energy consumption^[130, 131]. For example, hydrogels are mainly formed by non-covalent interactions between PRO and POL, while covalent interactions are less common^[81].

Among non-covalent forces, electrostatic interactions represent a primary driving force for PPC formation, given their sensitivity to pH, ionic strength, and biopolymer ratios^[103, 115, 117]. Hydrophobic interactions play an essential role in the stabilization of hydrophobic domains, particularly during encapsulation of hydrophobic BCs. Hydrogen bonds demonstrate direction and saturation characteristics, thereby enhancing the intermolecular bonding network. Van der Waals forces comprise dipole molecular interactions; although individually weak, their cumulative contribution significantly preserves structural integrity and conformational stability. In addition, π - π stacking can occur; these interactions are commonly classified as either hydrophobic or aromatic ring interactions. These non-covalent interactions typically exert synergistic effects through precise regulation of PPCs formation, architectural organization, and functional properties^[23, 126]. In this context, antisolvent precipitation has emerged as a particularly promising approach for encapsulating hydrophobic BCs. Alternatively, environmental triggers such as pH shifting or heat treatment can be employed to promote protein conformational rearrangements, thereby reinforcing the internal network structure. The subsequent sections will systematically review research progress on anti-solvent precipitation, the pH-driven method, and thermal-induced method.

3.2.1 Antisolvent precipitation

Antisolvent precipitation, also known as desolvation or liquid-liquid dispersion technology, is a widely employed method for constructing PPCs^[132]. Particle assembly during antisolvent precipitation is

fundamentally driven by intermolecular force imbalances across solute, solvent, and antisolvent phases. The process involves generation of supersaturation, nucleation, crystal growth, and coagulation (Fig. 2D)^[56, 133]. Water, organic solvents, and supercritical CO₂ are commonly used as solvent-antisolvent phases. This process is typically performed at room temperature and does not require special equipment or complex operating conditions, making it suitable for thermosensitive BCs encapsulation^[56]. In this process, hydrophobic PRO (e.g., zein, gliadin) forms the core structure through hydrophobic interactions, while POL functions as a shell layer, stabilizing the particles via electrostatic interactions^[132]. Based on the above theory, two studies utilized antisolvent precipitation to fabricate zein-hydrophilic polysaccharide nanoparticles, achieving effective encapsulation of BCs and enhancing stability^[56, 134]. The particles fabricated by this method usually exhibit well-defined spherical morphology, relatively narrow size distribution, and good uniformity^[56, 132]. However, the use of organic solvents like ethanol introduces additional purification steps and costs, limiting their application in solvent-free food products. Besides, nanoparticles generated through antisolvent precipitation frequently suffer from subsequent coagulation, directly compromising size distribution uniformity. Therefore, introducing effective stabilizers during manufacturing is essential.

3.2.2 The pH-driven method

The pH-driven approach exploits protonation-deprotonation equilibria of ionizable biopolymer groups through controlled pH adjustment (Fig. 2E)^[132, 135]. Functional groups of biopolymers undergo protonation or deprotonation in various pH environments, which in turn changes the charge states and intermolecular interaction forces of the molecules^[136]. The pH-driven method is applicable to compounds with high solubility and stability in alkaline solutions^[136]. For instance, this technique is commonly applied to zein, a widely used hydrophobic protein that is unstable at acidic pH or under high ionic strength, for nanoparticle fabrication^[137]. The pH-driven method was used for preparing zein-caseinate-alginate nanoparticles to encapsulate propolis, which indicated alginate molecules readily adsorb onto binary zein-caseinate surfaces through electrostatic complexation. This polymeric coating significantly enhanced colloidal stability by preventing severe nanoparticle aggregation under highly acidic and gastrointestinal conditions^[138]. Compared with antisolvent precipitation, nanoparticles prepared by the pH-driven method offer remarkable advantages, including well-defined spherical morphology, narrow particle size distribution, higher encapsulation efficiency and loading capacity, and enhanced pH and salt stability^[132]. Additionally, the pH-driven method is characterized by operational simplicity, high efficiency, no organic reagents, minimal equipment, and energy requirement^[139]. However, a significant drawback of the pH-driven method lies in its extreme sensitivity to external conditions, which frequently results in limited structural adaptability within complex food matrices and gastrointestinal environments. Although this method is mainly used to prepare PPCs, it has also been extended to the fabrication of certain PPCOs.

3.2.3 Thermal-induced

Thermal-induced method relies on the heat-induced denaturation and aggregation of PRO^[25]. Upon heating, PRO unfolds and interacts with POL through hydrophobic forces, electrostatic interactions, and entanglements, forming 3D networks (Fig. 2F). Bora et al.^[140] developed a novel composite gel through thermal-induced gelation of *Momordica charantia* bioactive polysaccharide and WPI. This gel encapsulated *L. acidophilus*, significantly improving its post-freeze-drying survival and storage stability. Similarly, thermal-induced SPI-*Tremella fuciformis* polysaccharide (TFP) composite gels exhibited concentration-dependent enhancement in rheological properties, texture characteristics, water-holding capacity, and thermal stability with the addition of TFP^[141]. Thermal-induced avoids the use of chemical reagents to achieve desired outcomes. However, excessive heat exposure may compromise the activity of BCs, necessitating careful optimization of both temperature and duration.

3.3 Assisted construction approaches

In addition to the aforementioned primary fabrication techniques of PPCOs/PPCs, emerging auxiliary methodologies, including electrospinning (ES), drying methods, pulsed electric field (PEF), ultrasound (US), microwave (MW), and high/ultra-high pressure (HP/UHP), have been widely adopted to optimize delivery performance of PRO-POL systems. These technologies offer novel strategies for precise structural engineering and functional enhancement of composite carrier architectures (Fig. 3).

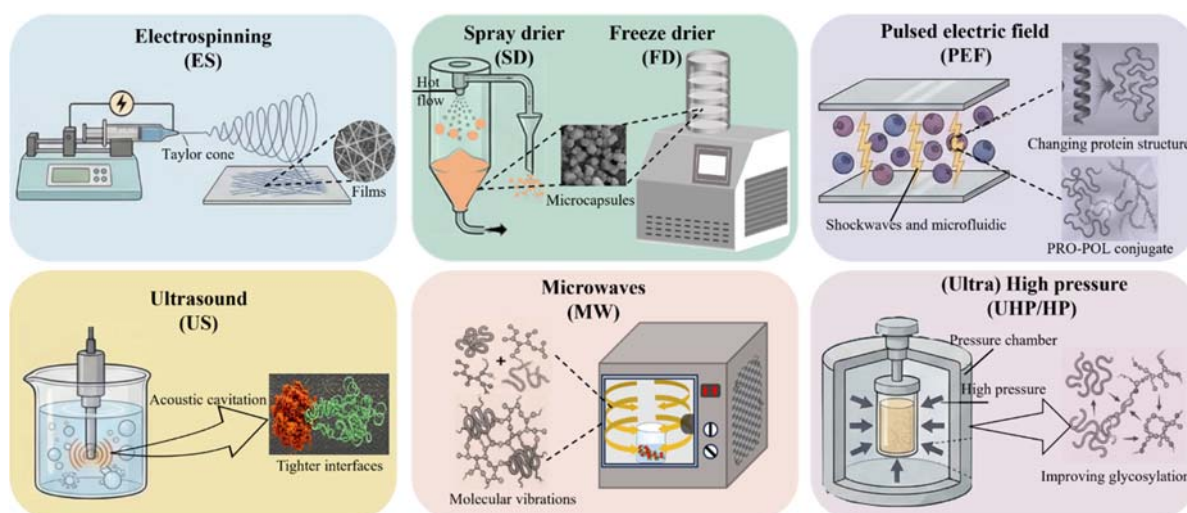


Fig. 3. Characteristics of assisted construction approaches.

3.3.1 ES

ES is an advanced, robust, and non-thermal processing technique primarily employed to fabricate functional nanofibers^[142]. Charged polymer fluids are ejected from a conical tip and travel toward a designated collector. Continuous electric field stretching and rapid solvent evaporation concurrently act upon these flying jets, driving final solidification into nanoscale fibers^[1, 143]. Ultrafine fibers were developed by adding fucoidan to renewable zein through ES, significantly enhancing the biological activity and functional performance of the material^[143]. PRO-POL-polyphenol bilayer composite membrane prepared by layer-by-layer ES showed superior performance in all aspects, which effectively delayed winter jujube

browning and prolonged shelf life^[94]. The resulting nanofibers exhibit distinctive properties, including minimal structural defects, enhanced mechanical properties, high specific surface area, porosity, permeability, and favorable biocompatibility^[144]. Coupled with ease of preparation and high loading capacity, ES holds great promise in the fields of food packaging, active ingredient encapsulation, and targeted delivery systems.

3.3.2 *Drying methods*

Among various drying methods, freeze drying (FD) and spray drying (SD) are the most commonly used. FD is a multi-stage process that includes freezing, primary drying (sublimation), and secondary drying (desorption), all carried out at low temperatures under vacuum. FD samples can be readily reconstituted and possess a long shelf life. However, the process demands high energy input, long drying times, and typically yields low throughput^[145]. It is suitable for encapsulating thermo-sensitive substances, like carotenoids and living cells, as it can minimize thermal degradation or inactivation^[146]. Obradovic et al.^[65] immobilized yeast cells in an inulin-WPI-maltodextrin carrier using FD, and powder exhibited microbial stability. In addition, FD typically produces samples with higher porosity, which may compromise the stability of encapsulated components and reduce embedding and retention efficiency^[147].

SD is a widely applied technology for the microencapsulation of food ingredients^[148], owing to its simplicity, rapidity, cost-effectiveness (30-50 times lower than FD), and high production capacity^[149]. The process typically employs aqueous dispersions, thereby minimizing the need for organic solvents, and generates microparticles with tailored composition, morphology, and microstructure for diverse applications^[150]. Similarly, SD is also commonly employed for loading BCs. Yang et al.^[149] developed soybean protein-soy polysaccharide complex via SD for encapsulating β -carotene, which possessed good stability in vivo, significantly reduced the degradation of β -carotene during long-term storage, and retained antibacterial activity. SD atomized emulsions into heated chambers, where rapid solvent vaporization generated dry particles encapsulating BCs. Conversely, lowering pressure after pre-freezing triggers sublimation of the solvent through a solid-to-gas phase transition, ultimately forming porous matrices enclosing BCs^[26]. However, the elevated temperatures used in SD may compromise the activity of thermosensitive compounds. Additionally, when combined with the Maillard reaction, the process forms reaction byproducts that influence the physicochemical properties of the final product^[26]. Currently, emerging drying technologies such as electrostatic spraying and ultrasonic drying offer innovative, energy-efficient routes for microcapsule production, broadening the scope for designing advanced delivery systems with tailored particle size, morphology, and functionality.

3.3.3 *PEF*

PEF, widely used in the liquid food for non-thermal pasteurization, is highly suitable as a pretreatment before drying, cooking, and freezing operations due to its low thermal load, reduced energy consumption, and environmental benefits^[151]. The main action mechanism of PEF is an electrical breakdown and

electroporation within the food matrix and associated microorganisms^[151]. PEF also induces transient shockwaves and microfluidic effects that alter protein structure, thereby yielding more stable emulsions with finer droplets and tighter interfaces^[152]. Recent studies have demonstrated the potential of PEF in the modulation of PPCOs and PPCs, and PEF strength might optimize the emulsifying properties of products^[153]. Taha et al.^[24] verified that PEF treatment promoted the Maillard reaction between BSA and starch, thereby enhancing the stability of the emulsion stabilized by the BSA-starch conjugate. The study further revealed varying electric field intensities exerted distinct impacts upon physicochemical properties of both conjugates and stabilized emulsions. These findings have been confirmed in related studies. The applications of PEF have successfully mitigated negative browning effects while improving inherent protein properties. Inducing glycosylation via glucose or mannose directly augments resulting foaming performance^[154]. Furthermore, PEF can improve the rheological properties of dough and liquids, thereby enhancing bread quality and color characteristics of liquid foods and increasing consumer appeal^[151]. In addition to its effects on interfacial properties, PEF has also been shown to induce structural modifications at the PRO level. PEF pretreatment of rapeseed PRO led to a significant reduction in α -helix content and a transition from ordered to disordered secondary structure, indicating that PEF directly alters PRO conformation and functional properties^[155].

3.3.4 US

US (20 to 1000 kHz) is an efficient (80-90%), economical and non-thermal food processing technology that can facilitate both PRO-POL interactions and Maillard conjugation^[156]. US influences polymerization following intensity- and time-dependent patterns. Low-frequency, high-intensity US (20-100 kHz, 10-1000 W/cm²) generates substantial mechanical energy and localized shear stress, actively inducing acoustic cavitation and microstreaming. Consequently, these dynamic micro-turbulences significantly enhanced overall thermal resistance, cryogenic stability, and inherent antioxidant potential across polymeric complexes, as well as increased the electrostatic and hydrophobic interactions to form compact complexes^[157, 158]. Conversely, lower power or prolonged exposure (e.g., 200 W or > 20 min) may increase aggregation due to the exposure of hydrophobic groups and the formation of disulfide bonds^[152]. Li et al.^[33] further demonstrated that appropriate US enhanced the electrostatic interactions between WPI-tremella polysaccharide systems, improving the flow characteristics of complex solution.

3.3.5 MW

MW involves electromagnetic waves with a frequency range between 300 MHz and 300 GHz, and offers rapid processing, environmental friendliness, operational simplicity, and scalability. MW generates molecular vibrations, which may lead to different PRO-POL interactions. MW-assisted Maillard reaction has been shown to improve solubility, emulsifying activity, and foaming ability of the resulting conjugates^[33]. Similar effects were documented in another study: MW accelerated the Maillard reaction between rice protein and dextran, enhancing rice protein solubility by approximately 40% and markedly increasing the grafting rate

compared with conventional heating^[159]. Furthermore, combined treatment with MW and PRO-POL nanoparticles for 5 min reduced the viability of *E. coli* and *S. aureus* by over 97%, demonstrating considerable potential for meat preservation^[95].

3.3.6 HP/UHP

HP, also known as cold pasteurization, utilizes non-thermal pressure to unfold PRO structures and promote molecular interactions, thereby forming ordered architectures and heat-stable gels^[160, 126]. HP is characterized by its high efficiency, low consumption, and safe operation, allowing maximal preservation of flavor compounds in food systems. During HP treatment, PRO experiences denaturation and conformational changes, including the disruption of electrostatic and hydrophobic interactions, and the changes in functionality, solubility, and digestibility^[161]. Similarly, POL undergoes pressure-induced structural alterations, leading to changes in molecular weight, binding capacity, viscosity, and other functional properties^[160]. Through HP treatment, the functional characteristics of myofibrillar protein were enhanced, and the biological activity of POL was maximized^[151]. Moderate pressure can enhance adsorption at the oil-water interface and improve film integrity, thereby strengthening emulsion stability and cohesion. However, excessive pressure may lead to aggregation or phase separation, weakening the emulsifier at the oil-water interface^[152].

UHP, an emerging green processing technology, utilizes water as a pressure-transmitting medium to apply up to 1000 MPa hydrostatic pressure to samples within sealed vessels^[162, 163]. UHP treatment significantly increase the rate and extent of glycosylation, yielding PRO with higher solubility, excellent emulsifying and foaming properties^[114]. In practical applications, foods treated with UHP suffer less damage to nutritional components and sensory quality^[162]. The integration of UHP with the Maillard reaction offers an efficient and sustainable strategy for constructing stable, functional, and tunable PRO-POL based delivery systems.

Overall, these assisted approaches effectively enhance encapsulation efficiency, antioxidant activity, emulsifying performance and structural tunability of PRO-POL based delivery systems. However, the functional performance of these systems is not exclusively determined by the preparation technique employed; it also depends on the complex interplay between structural characteristics and environmental parameters. Therefore, a systematic clarification of these influencing factors is essential for the rational design and targeted regulation of PRO-POL based carriers.

3.4 Factors affecting the performance of PPCOs/PPCs delivery systems

The physicochemical stability of PPCOs and PPCs is precisely maintained by covalent and non-covalent interactions. However, these forces are not indestructible and are susceptible to various influencing factors. This section systematically elucidates the factors influencing the delivery properties of PPCOs and PPCs from two primary perspectives, including the characteristics of wall materials (e.g.,

biopolymer type, mixing ratio, and concentration), preparation process (e.g., cross-linking methods and physical/chemical parameters).

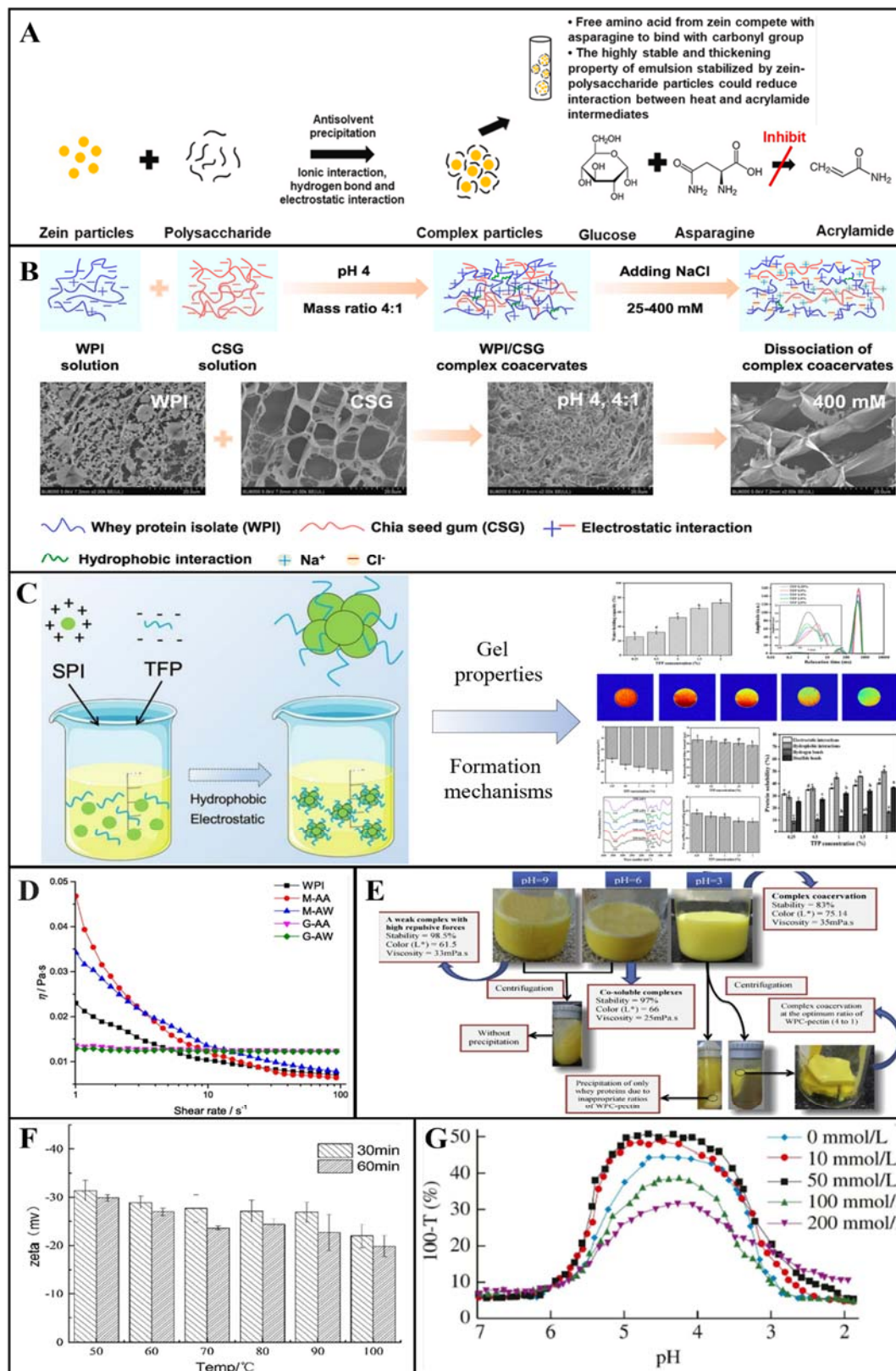


Fig. 4. Factors influencing the properties of PPCOs and PPCs. (A) Inhibitory effect of zein-POL microparticles on acrylamide^[164]; (B) Effect of mixing ratio between chia seed gum and WPI^[165]; (C) Impact of POL content on *Tremella fuciformis* polysaccharide-SPI gels^[141]; (D) Flow curve of the emulsions stabilized by M-AW and G-AW^[166]; (E) Influence of pH on the formation and stability of WPC-pectin complex^[167]; (F) Effect of thermal treatment on GA-zein-Cur suspension stability^[168]; (G) Turbidity curves of SPI/FVP solution with different NaCl concentrations^[169];

3.4.1 The characteristics of wall materials

The properties of biopolymers, such as PRO molecular weight and amphiphilicity, markedly influence the assembly and complexation of composite systems. The nature of PRO-POL interactions determines stability and environmental resistance of the resulting complexes. Generally, PRO can be classified into globular (folded state) and random coil (unfolded state). Globular proteins, such as BSA and soybean protein, tend to unfold under heat, variable pH, or ionic conditions^[23]. In contrast, random coil proteins exhibit intrinsically unfolded structures and react more readily with POLs. Certain POL, such as GA, soluble soybean polysaccharides, and sugar beet pectin, are natural PRO-POL conjugates with surface-active properties^[23], enabling modulation of heterogeneity and structural properties^[13]. The incorporation of POL with PRO can enhance the protective effects, encapsulation efficiency, and bioaccessibility of delivery systems^[23]. Therefore, the properties of the biopolymers must be fully considered when constructing delivery systems.

3.4.1.1 PRO/POL type

Under identical conditions, the choice of PRO and POL types markedly influences the physicochemical behavior of resulting conjugates or complexes. Champrasert et al.^[164] found that zein-POL composite particles could reduce acrylamide levels, with the most effective compound being zein-alginates, followed by zein-pectin (Fig. 4A). Meanwhile, modified yeast protein-CS containing *Monascus* pigments exhibited stronger hydrogen bond interactions, leading to better encapsulation and improved color retention and storage stability. Modified yeast protein-XG based system displayed higher physical stability and enhanced elasticity^[37]. Another study demonstrated that gelatin combined with high-methoxyl pectin or cashew gum showed superior specific lipid-binding capacity compared with other POLs^[170]. This conclusion holds across different protein matrices as well. Al-Maqtari et al.^[68] investigated three PROs, namely WPI, SC, and SPI, in combination with GA to encapsulate *Pulicaria jaubertii* extract (PJ), in which SC-GA-MPJE exhibited a smooth surface feature, no visible cracks, as well as the most excellent performance. The stability and delivery efficiency of PPCs and PPCOs are governed by molecular interactions. Insufficient POL charge density or PRO structural sensitivity may trigger dissociation or leakage of encapsulated compounds. Therefore, precise matching of components is essential to ensure optimal performance.

3.4.1.2 PRO/POL mixing ratio

The mixing ratio of PRO to POL has a significant effect on the morphology and stability of conjugates or complexes. Insufficient POL content weakens electrostatic repulsion, leading to particle aggregation. As the POL ratio increases, the particle size of nanoparticles decreased gradually and the thermal stability improved^[47]. The mixing ratio also dictates the type of structures formed, where low POL levels favor nanoparticle formation, whereas high POL levels produce network-like microgels^[14]. L. Li et al.^[165] demonstrated that when the mixing ratio of WPI to chia seed gum (CSG) was adjusted to 4:1, the resulting WPI-CSG coacervates exhibited the strongest electrostatic interactions (Fig. 4B). Similarly, a psyllium seed

gum (PSG)-to-WPI ratio of 1:1 yielded composite films with the highest tensile strength and elastic modulus, along with optimal overall physicochemical and mechanical properties. At higher PSG concentrations, the composite film showed reduced color variation, as well as higher brightness and whiteness^[171].

3.4.1.3 PRO/POL concentration

The complexes formed by PRO and POL at varying concentrations exhibited significant differences in the loading efficiency of BCs, physicochemical stability, and structural characteristics of composites. In a rice protein-pectin composite gel, a pectin concentration of 1.5% enhanced water-holding and Cur encapsulation efficiency, loading capacity, and cumulative release^[172]. Incorporating *Dendrobium officinale* polysaccharide (DOP) enhanced the thermal stability of WPI-DOP gels. At a DOP concentration of 1.5%, the gel network became more compact, homogeneous, and stable^[173]. Increasing the concentration of Tremella fuciformis polysaccharide (TFP) enhanced non-covalent interaction between SPI and TFP (Fig. 4C)^[174]. In PRO-POL biopolymer particles, the inhibitory effect of zein on acrylamide was exerted in a dose-dependent manner, with high concentrations unexpectedly promoting acrylamide formation^[164].

The selection of appropriate biopolymer types, mixing ratios, and concentrations is critical for tailoring delivery systems with desired particle size, hydrophilicity, surface charge, and release profiles. Therefore, in the combination of PRO and POL, the wall materials should be correctly and reasonably selected so as to achieve the desired performance of PPCOs/PPCs.

3.4.2 Preparation process

3.4.2.1 Cross-linking methods

Both physical blending and chemical cross-linking influence the structure and stability of PPCs and PPCOs. Physical methods such as thermal-induced and pH-driven offer favorable biocompatibility, but yield relatively lower stability compared with covalent strategies. A previous study comparing physical mixtures with Maillard-derived covalent conjugates revealed that the Maillard reaction conferred substantial advantages in droplet size, storage stability, and bioavailability of functional factors^[14]. Ai et al.^[166] compared Maillard-crosslinked WPI and sugar beet pulp polysaccharide emulsions (M-AW) with genipin-crosslinked systems (G-AW), finding that despite lower electrostatic repulsion, M-AW maintained comparable or even emulsion stability due to high viscosity (Fig. 4D). This experiment confirmed that specific cross-linking sites have a critical impact on PPCOs.

Although both physical and chemical cross-linking strategies present unique advantages and inherent limitations, comparative investigations of PRO-POL delivery systems fabricated through distinct cross-linking approaches remain noticeably limited, necessitating comprehensive mechanistic investigations.

3.4.2.2 Physical and chemical parameters

pH critically governs electrostatic complexation by modulating molecular charge and structure, thereby dictating molecular interaction strength and structural properties^[175]. Alkaline pH favors the Maillard

reaction, further affecting cross-linking efficiency^[175]. At the pI, PRO is electrically neutral, displays minimal solubility, and tends to aggregate. Under acidic pH conditions, PRO tends to denature, unfold, and expose more active sites^[4]. When pH is lower than PRO's pI but remains above the pKa of the anionic POL, the positively charged PRO will form a complex with the negatively charged POL through enhanced electrostatic attraction^[176]. Further acidification intensifies this attraction until charge neutralization yields an unstable complex that lacks sufficient electrostatic repulsion, ultimately leading to phase separation and increased turbidity^[165, 177]. However, if the pH continues to decrease further below the pKa of POL, protonation of carboxyl groups reduces the negative charge, thereby weakening the electrostatic interaction with PRO and leading to coacervate dissociation^[178]. Conversely, when pH exceeds the pI, PRO may carry a partial positive charge or a net negative charge, resulting in either electrostatic attraction or repulsion with POL, which can disrupt network formation or induce phase separation^[176, 179]. Ghasemi et al.^[167] demonstrated that the stability of WPI-pectin systems varies significantly across different pH levels. Specifically, the stability was minimal at pH 3.0 and presented complex coacervation; whereas at pH 9.0, the maximal stability and high repulsive forces were reached (Fig. 4E). Notably, excessive reliance on pH-responsive designs tailored to specific pH ranges may result in unpredictable behavior under complex physiological conditions, compromising targeting efficacy.

Temperature plays a crucial role in modulating molecular mobility and the strength of intermolecular interactions, which distinctly impact the assembly and structural integrity of PRO-POL delivery systems. Both elevated and reduced temperatures may disrupt complex structures or lead to inhomogeneous cross-linking. Increasing temperature strengthens hydrophobic interactions by enhancing molecular mobility, whereas lower temperatures favor the formation and stabilization of hydrogen-bond networks^[176]. In addition, thermal treatment affects physical, structural, and morphological characteristics of PRO^[180]. Notably, extreme temperatures may cause PRO and POL denaturation or degradation. In zein-GA core-shell nanoparticles designed for Cur delivery, exposure to temperatures above 70°C altered the spatial structure of zein, causing partial dissociation of GA from the PRO surface, reduced surface charge, and ultimately triggered Cur degradation (Fig. 4F)^[168]. These findings demonstrated that extreme temperatures can significantly alter the structural characteristics, thereby affecting the delivery efficacy. Additionally, the delivery systems formed under different temperatures exhibited distinct phase states. Hydrogels prepared from BSA and gelatin responded differently to temperature. At 10°C, the hydrogel formed a continuous gelatin network with dispersed BSA inclusions, whereas at 80°C, BSA constituted the continuous phase, with liquid gelatin dispersed as inclusions within it^[181]. These findings underscore the importance of optimizing temperature to balance structural integrity and encapsulation efficiency during processing. Furthermore, certain studies have developed multi-pH- and temperature-responsive systems that utilize these environmental factors. These systems enable encapsulated polyphenols within nanocarriers to maintain stability under gastric conditions while rapidly degrading in localized alkaline environments characterized by elevated temperatures^[182].

Ionic strength critically modulates the electrostatic interaction that governs the stability and assembly behavior of PRO-POL based delivery systems. Zhang et al.^[183] demonstrated that NaCl competed with positively charged PRO and negatively charged POL for binding sites, thereby reducing the probability for complex formation. Similarly, Nasrabadi et al.^[4] found that emulsion instability at high salt concentrations was primarily attributed to the charge screening effect of salts and unfavorable conformational alterations in PRO constituents. This effect attenuates both electrostatic attraction and repulsion, partially impeding the self-assembly process^[184]. Additionally, high salt ions were verified to inhibit PRO protonation by shielding H^+ ions, thereby reducing dissociation and system turbidity^[185]. Conversely, neutral salts at lower concentrations promote charge neutralization, enabling the formation of compact PRO-POL aggregates (Fig. 4G)^[169]. However, excessive ionic strength can disrupt the balance of electrostatic attraction and repulsion between PRO and POL, drastically lowering the critical pH and completely inhibiting complexation^[165, 185]. Moreover, differences in ion types and valency also exert diverse effects on the PRO-POL delivery systems. Divalent ions (like Ca^{2+} and Mg^{2+}) can form ionic bridges with carboxyl and sulfate groups on both PRO and POL, thereby restructuring the network and impairing delivery performance. Compared to monovalent cations, multivalent cations suppress electrostatic attraction more effectively and inhibit complexation at lower ionic strengths^[176].

Current research should give sufficient attention to the aforementioned influencing factors. Existing investigations into oxidative environment effects on delivery systems predominantly focus on characterizing thiol group oxidation. The regulatory mechanisms governing aggregate formation through process parameters such as mechanical stirring remain incompletely understood, while the underlying principles of auxiliary treatments including US and FD are still largely phenomenological. Consequently, the practical efficacy of these systems in applications, including BCs delivery, texture modification, and nutritional enhancement, remains to be established. Further work is needed to elucidate the relationship between influencing factors and delivery systems, along with validating their efficacy within real food matrices. The interplay between molecular interactions and environmental conditions that determines PRO-POL complex assembly is summarized in Fig. 5.

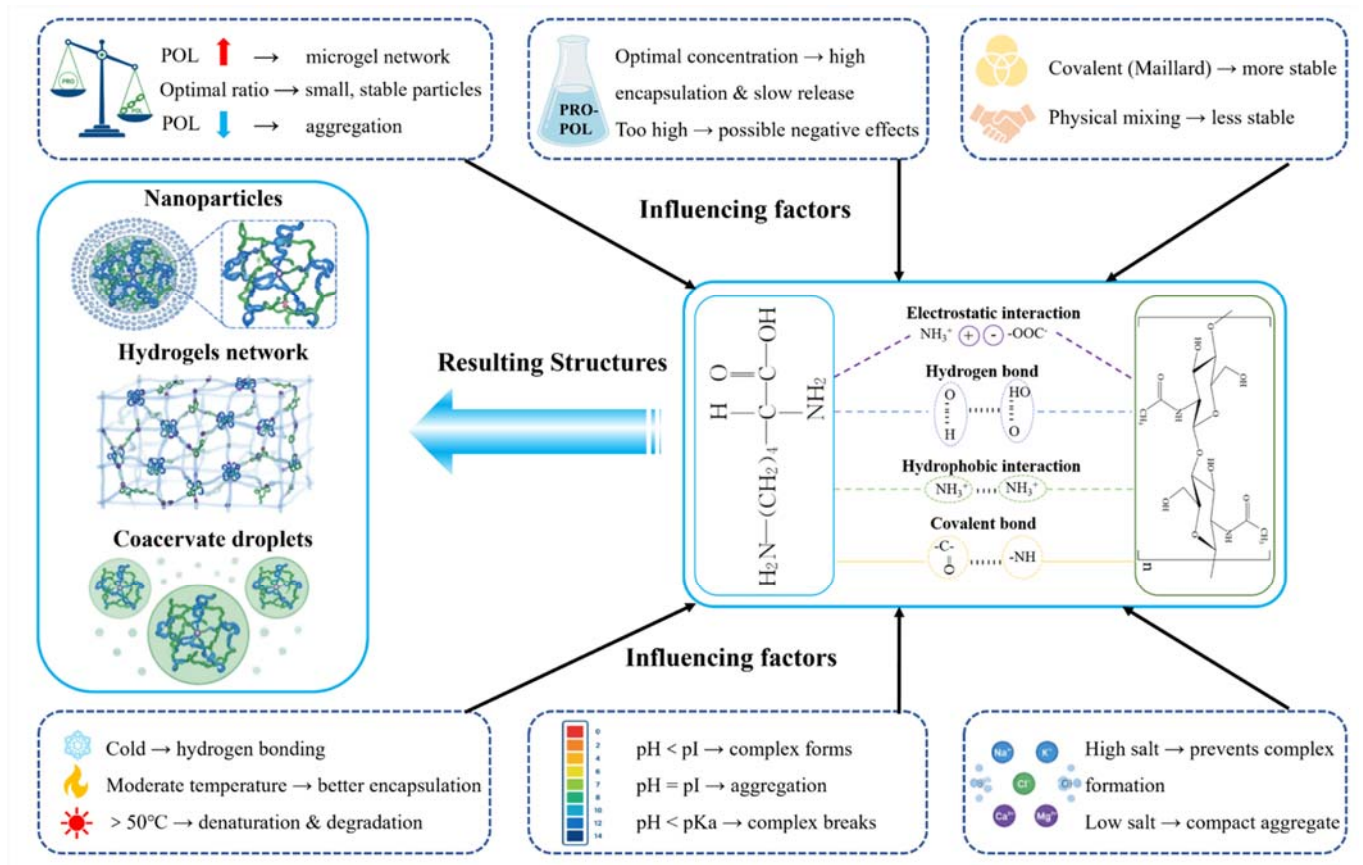


Fig. 5. PRO-POL interaction mechanisms and regulatory network of influencing factors

4. Applications in the food industry

The preceding sections have clarified how biopolymer properties, preparation methods, and environmental factors govern the formation and functional performance of PPCs and PPCOs. This section turns to their translational applications. These mechanistic insights enable the rational design of delivery systems tailored for specific food-industry needs. Such applications include enhancing probiotic viability, fortifying nutrients, developing active packaging, replacing lipids, and enabling advanced manufacturing techniques such as 3D printing. The applications of PPCOs/PPCs in the food industry are illustrated in Fig. 6.

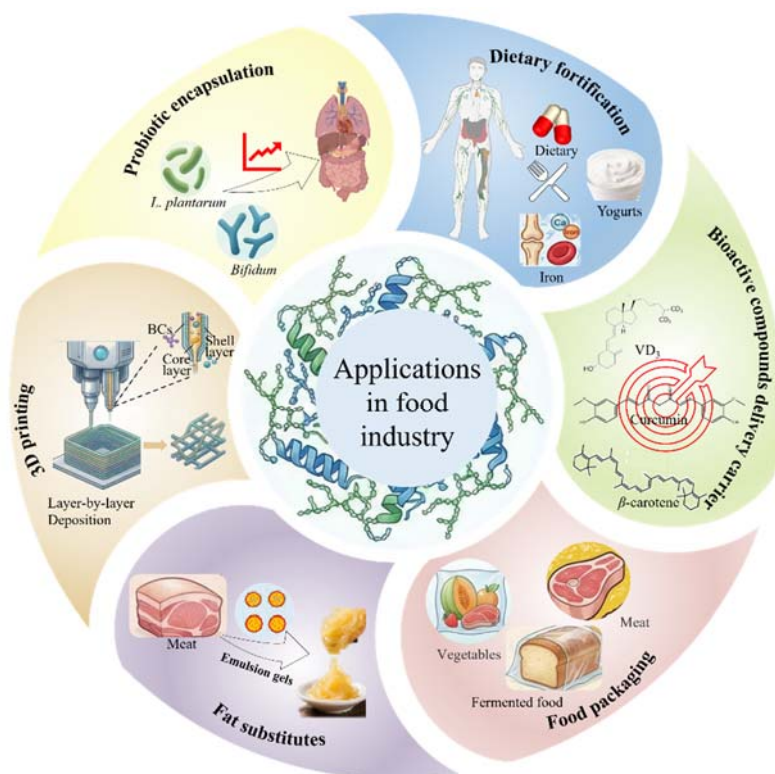


Fig. 6. Overview of PPCOs/PPCs applications in the food industry.

4.1 Probiotic encapsulation

Probiotics can modulate and promote intestinal microbiota health and are therefore widely used to support gut health. Numerous studies have confirmed that PRO-POL based delivery systems can effectively improve the survival rate and targeted delivery of probiotic. For example, a transparent edible film composed of WPI and kefir polysaccharide actively protected *Kluyveromyces marxianus* CIDCA 8154 and *L. paracasei* CIDCA 8339 during drying and simulated digestion, maintaining high cell viability^[97]. Similarly, *L. plantarum* strains encapsulated by casein-CS microparticles^[186] or WPI-dextran^[187] exhibited improved survival rate and functionality under storage conditions. In addition, cross-linking casein-CS microparticles with appropriate agents was examined to enhance the survival of probiotics. Krunić & Rakin^[188] demonstrated that the beverage formulated with a WPI-alginate gel positively influenced both the antioxidant properties and the viability of probiotic during 28-day storage. Furthermore, protein-alginate beads encapsulating *Streptococcus thermophiles*, *L. delbrueckii ssp. Bulgaricus*, *L. acidophilus*, and *Bifidobacterium bifidum* maintained over 96% viability rate under gastrointestinal conditions, significantly exceeding that of free cultures^[189]. The two aforementioned studies used proteins and their hydrolysates as carriers for probiotics delivery. Their findings align with the observations in Section 3.4.1 of this review regarding the influence of different wall materials on delivery system. Additionally, Zhu et al.^[38] verified that the viability of *L. plantarum* under gastrointestinal conditions was remarkably enhanced by encapsulating it within C-phycocyanin-pectin hydrogels. Overall, these findings indicated that encapsulation significantly enhanced

probiotic survival and improved economic efficiency, and that higher protein content increased the hydrogel's hardness, strength, and stability.

4.2 Dietary fortification

Dietary fortification aims to enhance the nutrient density of daily food by incorporating essential vitamins and minerals, thereby meeting specific population needs and preventing micronutrient deficiencies. For example, walnut meal protein hydrolysates (WMPHs) and chitosan oligosaccharides (COS) were used to prepare glycosylated peptide-calcium chelate (WMPHs-COS-Ca) via the Maillard reaction, the effects of temperature and pH on the saccharification and browning of WMPHs-COS-Ca also be studied^[190]. The resulting WMPHs-COS-Ca showed excellent stability and bioavailability under high temperature, various pH conditions, and gastrointestinal tract environments, while providing controlled calcium release favorable for absorption. For iron fortification, a WPI-GG based cold-set hydrogel achieved 94% encapsulation efficiency and released iron predominantly during the simulated gastric phase rather than the intestinal phase^[191]. Another study verified that WPI-GA nanoparticle slowed iron release to 20% under intestinal-simulated digestion^[192]. These studies provided valuable research insights and data support for further optimizing minerals delivery systems and improving effectiveness in dietary fortification applications.

Numerous studies also systematically explored the application of PRO-POL delivery systems in vitamin supplementation. VD₃ was encapsulated in gelatin-maltodextrin and whey protein concentrate-pectin, respectively^[193, 58]. In addition, the concentrations of wall material and surfactant were also investigated to achieve optimal encapsulation. Fortification of yogurts with encapsulated VD₃ led to improved textural properties, overall acceptability, and probiotic viability of yogurt. The latter effectively fortified sunflower oil by enhancing bioavailability and shelf life. The microcapsule prepared with amaranth carboxymethyl starch and LF enabled the incorporation of β -carotene into gummy candies, improving the candies textural properties^[194]. It exhibited favorable β -carotene bioaccessibility and was used to supplement high-risk populations with vitamin deficiency. The aforementioned studies demonstrated that encapsulating vitamins within PRO-POL delivery systems not only enhances nutritional fortification but also effectively addresses vitamin deficiency disorders.

In addition to incorporating supplementary substances, PRO-POL delivery systems can also stabilize endogenous food components, thereby achieving dietary fortification. For instance, the colloidal particles of Pickering emulsion based on α -lactalbumin and CS exhibited protective effects against oxidation in menhaden oil^[195]. The film of α -lactalbumin-CS colloidal nanoparticles had higher surface hydrophobicity than α -lactalbumin at pH 5.0, and 6.5. Polymerized WPI-dendrobium officinale polysaccharide gel enhanced the quality of goat milk yogurt by improving the particle size, dehydration shrinkage, and viscosity^[173].

4.3 BCs delivery carrier

Encapsulation of BCs (e.g., polyphenols, vitamins, and carotenoids) via PRO-POL systems can improve the stability, solubility, and bioavailability, holding broad prospects for future development. Cur was encapsulated by pea protein isolate-pectin complexes^[73], and WPI-HA nanoparticles^[43], which all showed stronger protection against thermal and photo degradation. Additionally, this study investigated the influence of preparation methods on complexes, demonstrating that variations were observed across different approaches. For ACNs, glycosylated casein-carboxymethyl cellulose encapsulation remarkably enhanced the chemical stability and achieved controlled release^[64]. The bioavailability of soybean isoflavones was enhanced by encapsulating them within SWP-SA emulsion system^[61].

The encapsulation of vitamins within PRO-POL delivery systems enables sustained release properties. CS-gelatin hybrid polymer networks containing VB₁- β -cyclodextrin were prepared, which achieved controlled release of VB₁^[196]. OVA-pectin or carboxymethyl tara gum-LF was synthesized to encapsulate VD₃, enhancing the stability and enabling controlled release^[197, 198]. Various POLs and zein-based microcapsules have been evaluated for their ability to stabilize VE, and zein-XG microcapsules in particular showed a high encapsulation rate for VE, along with excellent thermal stability and sustained-release properties^[8].

For carotenoids, *Flammulina velutipes* protein-polysaccharide^[10], WPI-GG^[199], or WPI-GA^[112] were prepared to load β -carotene, all of which showed higher encapsulation efficiency, enhanced stability, and bioavailability. Chu et al.^[11] fabricated hydrogels from SPI and soybean oligosaccharide, which had higher photochemical stability and bioavailability of zeaxanthin, as well as slower chemical degradation. Astaxanthin was successfully encapsulated by HPP-CS Pickering emulsion^[20] and myofibrillar protein-CS emulsion^[63]. The former demonstrated superior sodium reduction capacity and antioxidant activity, while the latter effectively prevented astaxanthin degradation during storage. Furthermore, the concentration of HPP-CS complex was positively correlated with the structural and thermal stability of Pickering emulsion. Collectively, these results demonstrated that BCs-loaded PRO-POL delivery systems exhibited superior physicochemical stability in food applications, significantly enhancing nutrient utilization efficiency.

4.4 Food packaging

Traditional coatings or films suffer from limitations, such as poor biocompatibility, inadequate degradability, and limited functionality, which restrict their application in food preservation. Recent advances have utilized PPCOs/PPCs to encapsulate BCs for developing edible coatings, demonstrating noticeable shelf life extension in perishable products, including fruits, meats, and vegetables, etc.

ES and layer-by-layer assembly techniques were employed to prepare zein-wheat gluten-CS-gelatin-thymol composite fiber film with superior thermal stability, mechanical properties, water vapor permeability, and antibacterial capability, which remarkably extended the shelf life of winter jujubes^[94]. Zein-shellac-thymol core-shell nanoparticles exhibited potent antimicrobial efficacy, effectively prolonging cantaloupe freshness by 16 days^[200]. Multifunctional films were fabricated by incorporating carbon dots into gelatin/*Sargassum fusiforme* polysaccharide matrix, demonstrating excellent antioxidant

activity and inhibitory efficacy against *S. aureus* and *E. coli*. The resulting films effectively preserved fruit quality when applied to blueberry packaging^[92].

PRO-POL based packaging materials also present considerable efficacy in meat preservation. Egg albumen-sodium carboxymethylcellulose^[95], or hydroxypropyl methylcellulose-pectin-gelatin^[201] was prepared as packaging films, which all exhibited excellent antibacterial performance and achieved meat preservation. Ben Azaza et al.^[202] fabricated an active packaging film based on CS and sardinella protein isolate, which enhanced the microbial stability and reduced the lipid peroxidation of wrapped shrimp during cold storage. The above examples illustrate the importance of composite films for meat preservation. Although PRO-POL films provide considerable preservation benefits, the standalone efficacy is constrained. Maximum functional performance can be achieved through integration with refrigeration, which synergistically enhances preservation outcomes.

In addition to being commonly used for fruits and meat products, PRO-POL packaging has also demonstrated versatility in other food categories. For instance, cheese shelf life is often limited by microbial activity. Pluta-Kubica et al.^[91] developed active furcellaran /WPI based films with white tea or yerba mate, extending the shelf life of fresh soft rennet-curd cheese. Cashew gum-gelatin-ferulic acid-perch essential oil was employed to fabricate composite films, which not only gave bread more harshness but also prolonged the shelf life by 3 days^[203]. Sarraf-Ov^[96] fabricated a bio-nanocomposite film based on gelatin-CS nanoliposomes encapsulating tobacco extracts. The resulting film showed significant improvements in barrier properties, thermal stability, mechanical strength, antioxidant activity, and antibacterial efficacy compared to the control film, thereby highlighting its ability to extend the shelf life of perishable food products. Collectively, these findings indicate that PRO-POL film systems effectively delay food spoilage, substantially reduce economic costs, and concurrently fulfill food safety and health requirements.

4.5 Fat substitutes

Fat substitutes contribute to reducing the incidence of chronic diseases (e.g., obesity and diabetes) associated with high-fat diets. However, fat reduction often leads to undesirable sensory characteristics. Therefore, emulsion gels have been developed as partial fat replacements that achieve comparable taste and texture profiles to conventional fats. When incorporated into bologna sausage, high-fiber emulsion gels prepared from inulin and SPI imparted a softer texture, higher elasticity, improved viscosity, and enhanced lipid oxidation stability. Emulsion gels effectively restored the matrix structure weakened by fat and salt reduction, while demonstrating superior fatty acid composition and stable refrigerated storage performance, establishing its potential as a healthy fat alternative^[204]. Öztürk-Kerimoglu^[205] systematically elucidated how a composite gel system comprising pea protein and agar modulates the functional properties, chemical characteristics, and nutritional profile of model meat emulsions, demonstrating its potential applicability in processed meat product development. Huang et al.^[206] drew a similar conclusion that a composite emulsion gel containing 1% SPI and 4% KGM exhibited friction coefficients and color parameters comparable to those of natural fat and more closely mimicked the swallowing sensation and lubricity of natural fat.

Emulsion gels prepared with SPI-curdan exhibited thermal-irreversible properties, and at oil content of 10 wt%, the gel showed comparable hardness, chewiness, springiness, and gel strength to pork backfat without evident differences^[207]. Moreover, the emulsion gels synthesized from soybean oil, SPI, agar, and alginate exhibited a dense network structure capable of entrapping oil droplets, indicating elastic behavior across all tested temperatures, and displayed superior hardness. Their textural properties closely resembled porcine adipose tissue, thereby significantly enhancing the quality of plant-based meat analogs^[49]. Overall, PRO-POL based emulsion gel serves as a functional fat analog, enabling substantial fat reduction in food products while maintaining organoleptic properties, thus facilitating the development of healthier food alternatives.

4.6 3D printing

3D printing is an advanced prototyping manufacturing method to produce complex structures by depositing materials layer-by-layer, integrating computing, precision transmission, and material science^[88, 89]. Due to the advantages of functional material design, personalized structural customization, and processing simplification, 3D printing has garnered remarkable attention in the food industry^[208, 209]. By constructing a multi-level protein-polysaccharide interaction network, the apparent viscosity of the composite system can be significantly increased, ensuring the dimensional accuracy and structural integrity of the printed objects^[59]. The yield stress generated by non-covalent and covalent cross-links supports successive layer deposition, and its magnitude and nature are governed by the type and concentration of the biopolymers. Therefore, appropriate selection of proteins and polysaccharides enables precise control over the ink viscoelasticity and mechanical strength to meet the requirements of the 3D printing process^[209].

3D printing ink rich in β -carotene was formulated from SPI, corn starch, and Ora-pro-nobis mucilage, exhibiting flexible structure, high printing accuracy, and strong mechanical properties^[59]. Building on this single-material approach, coaxial 3D printing was further employed to fabricate an inner-core gel, in which ACNs and Cur were co-encapsulated within an SPI-starch-XG stabilized emulsion. This more complex structural design conferred a strongly bound water state and excellent storage stability, thereby markedly improving the retention of both ACNs and Cur during long-term storage^[88]. Beyond emulsion-based systems, a MCT oleogel structured with GG and gelatin was developed as an alternative co-delivery carrier for Cur and resveratrol. Custom-shaped 3D printing of this oleogel system not only enhanced the oral bioavailability of the encapsulated bioactives but also enabled intestinal-targeted delivery, further expanding the functional versatility of 3D-printed PRO-POL delivery platforms^[55].

PRO-POL delivery systems can also be realized through 3D printing of probiotic-loaded constructs. Probiotic encapsulations were fabricated from WPI, maltodextrin and fructooligosaccharide via combined spray freeze-drying and 3D printing, and their effect on probiotic stability was evaluated under two storage conditions (4°C and room temperature)^[210]. When tea protein XG-Pickering emulsion gel was used as the base material for 3D printing inks, it was found that a printing temperature of 65°C significantly reduced the

number of live probiotics^[211]. These innovations lay the foundation for 3D printing in developing personalized nutritional health products.

Beyond 3D printing, 4D printing introduces dynamic self-assembly capabilities, enabling materials to respond to external stimuli, including temperature, humidity, or pH changes. This allows for more precise controlled release and locations^[212, 213]. Advancements in PRO-POL based delivery systems are anticipated to drive developments in 4D printing, offering transformative potential for precision nutrient delivery and functional food design.

4.7 Other food-related applications

The potential applications of PRO-POL complexes extend well beyond the aforementioned fields of food processing and nutritional health, now encompassing numerous additional domains. PPCOs and PPCs have prominent potential in inhibiting acrylamide formation during heat treatment, aligning with the trend towards clean-label food while maintaining desirable texture. Zein-SA and zein-pectin nanoparticles effectively inhibited the formation of acrylamide in treated French fries^[164]. Hollow salt particles stabilized by PRO-POL complexes within emulsions may serve as a potential low-salt alternative and dietary supplement. At equivalent mass, the saltiness of the hollow salt was comparable to that of conventional NaCl, while enabling reduced sodium intake. Meanwhile, higher astaxanthin retention and oxidative stability were observed in the hollow salt during storage at 50°C for 15 days^[20]. Beyond these applications, PRO-POL delivery systems indicated the ability to delay gastric emptying to prolong the sensation of fullness, and they can be engineered into palatable food products for patients with dysphagia^[214]. These findings indicated their possible utility in weight management and medical foods. In summary, PRO-POL complexes have evolved from simple carriers of BCs into a versatile food design platform capable of performing multiple functions, including inhibiting processing-induced harmful substances, reducing salt and enhancing nutrition.

5. Limitations

Although PRO-POL based delivery systems offer considerable advantages in stability, controlled release characteristics, and bioaccessibility enhancement, they still present several challenges in practical applications. Firstly, it is difficult to optimize multiple functional properties such as high loading capacity, strong protection, controlled release and high bioaccessibility and bioavailability simultaneously, as most current systems prioritize only one or two parameters^[103, 215]. Then, PRO-POL systems exhibit pronounced sensitivity to environmental variables^[216], parameters optimized in the laboratory often fail in real food matrices due to competitive effects from sugars, salts, and lipids^[47]. The inherent structural complexity and source variability of natural BCs further compromise reproducibility and scalability^[215]. Finally, current research predominantly focuses on in vitro studies, without systematic animal or clinical trial data to support safety and efficacy. Moreover, cross-linking agents introduce process complexity through required residue

removal steps, creating potential toxicological risks^[47]. This creates a critical disconnect between laboratory-scale feasibility and the rigorous safety certifications mandated for industrial applications^[44].

6. Future development trends

PRO-POL based delivery systems will advance toward intelligentization, functionalization, and sustainability in the future, while their practical applications will be further propelled.

6.1 Intelligent prediction

Artificial intelligence (AI) is emerging as a powerful tool for designing novel PRO structures and predicting PRO-POL interactions^[99, 217]. Pillai et al.^[218] successfully utilized AI to design novel PRO structures capable of precisely regulating assembly or disassembly through conformational changes. AI training simulations necessitate extensive datasets. While numerous PRO-POL delivery systems have been documented, most employ identical materials interchangeably, resulting in insufficient data diversity to support robust AI simulations. Consequently, there exists an imperative to synthesize novel PROs and POLs that enhance diversity within dataset construction. Additionally, multi-scale learning enables mechanistic simulations and predicts complex formation based on binding energy, sites, and interactions in molecular docking^[21]. Molecular dynamics simulations analyze binding processes and conformational changes at the atomic scale. The advancement of intelligent simulation will become a major trend moving forward.

6.2 Functional foods and targeted delivery

In the food sector, growing demands for functional health foods have highlighted the value of PPCOs and PPCs in disease-specific nutritional interventions. Recent studies confirm their suitability for developing personalized functional foods targeting patients with diabetes, obesity and cardiovascular diseases^[25]. However, the research and development of functional foods extends beyond preparation processes; safety assessment is equally critical and must include both in vitro and in vivo evaluations^[119]. At present, most studies still focus on in vitro safety evaluation. Obviously, differences exist between in vitro and in vivo environments due to variations in physiological conditions and other complex factors, meaning results from the two approaches may not always align. For example, studies have found that although certain gel materials are rapidly absorbed by the human body, they cannot be effectively broken down or digested^[25]. Systematically investigating the fate of delivery systems in the digestive tract and throughout systemic circulation can provide both theoretical foundations and practical guidance for designing smarter, more efficient nutritional supplements and BCs delivery systems^[132, 219].

6.3 Large-scale production

The structural and compositional complexity inherent to lab-scale fabrication of protein-polysaccharide nanoparticles inevitably increases production costs^[119]. Therefore, the development of cost-effective, scalable composite systems is critical^[220, 221]. For industrial implementation, automated manufacturing platforms and AI-driven process optimization should be prioritized to mitigate human errors and enhance

reproducibility. Interdisciplinary collaboration can facilitate novel applications of biopolymers in medical and environmental sectors^[46]. Prior to practical deployment of optimized nanocomposites, comprehensive characterization of their performance parameters must accompany advancements in industrial production technologies. These parameters include sensory attributes, allergenic potential, physicochemical stability, bioavailability profiles, BCs loading efficiency, controlled release, and targeted delivery.

6.4 Sustainable development

From an ecological perspective, PPCOs and PPCs derived from renewable sources like plants, animals^[202, 101], and microorganisms^[57, 92] reduce environmental footprint during production, as well as dual advantages of cost-effectiveness and eco-friendliness, perfectly aligning with the development requirements of green food industry^[46, 217]. In addition, the development of low-temperature, low-pressure, and low-energy preparation methods, such as employing mild techniques to replace traditional chemical crosslinking, can significantly reduce the use of chemical reagents, thereby minimize environmental pollution and preserve the structural integrity of BCs during production. Finally, PPCOs/PPCs can integrate multiple smart functions, especially intelligent detection, to enhance material utilization efficiency and support sustainable development.

7. Conclusion

PRO-POL based delivery systems have emerged as promising platforms in the food industry, offering distinct advantages in terms of biosafety, cost-effectiveness, and functional benefits. This article systematically reviews the commonly used PRO/POL raw materials and types for delivery systems. Based on different interaction forces, the preparation methods and auxiliary techniques for PPCOs and PPCs are introduced, together with the key factors influencing their delivery performance. In addition, the current applications in the food industry are comprehensively reviewed. However, the system's toxicological properties and sensitivity to environmental variables warrant further investigation. These delivery systems are anticipated to expand into domains including functional foods, targeted delivery, sustainability, and smart enhancement in the future. Moreover, AI-driven analytical approaches may offer enhanced understanding of PRO-POL fundamental interaction mechanisms. PRO-POL delivery systems have attracted considerable attention from both researchers and food industry, and their design and development are becoming increasingly mature. Foundational studies systematically elucidated the composition and formation mechanisms of PPCs and PPCOs, offering a critical basis for the rational design of BCs delivery systems. By enabling food preservation, nutritional fortification, and the safe transport of bioactive agents, these systems directly respond to the growing societal demand for health-promoting foods, thereby driving innovation in future food technology and guiding the deliberate design of diverse nutritional and health product delivery systems.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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