

Condensed Matter and Interphases

ISSN 1606-867X (Print)

Kondensirovannye Sredy i Mezhfaznye Granitsy https://journals.vsu.ru/kcmf/

Review

Review article https://doi.org/10.17308/kcmf.2024.26/11933

Stabilization of food emulsion by polysaccharides and protein-polysaccharide complexes: a short review

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Abstract

Emulsions are heterogeneous systems consisting of two immiscible liquids, widely used in the food industry as the basis for some products (mayonnaise, sauces, etc.) and components for the production of functional food products containing systems for targeted delivery of biologically active substances (vitamins, nutraceuticals, flavonoids, etc.). From a thermodynamic point of view, emulsions are unstable systems with excessive surface energy; therefore, they are characterized by rapid destruction through phase separation. For the solution to this problem, emulsifiers are used, amphiphilic molecules of various natures that reduce surface tension, i.e., possess surface activity. However, most of these stabilizers are synthetic and toxic products, which significantly limits their use in the food industry. Natural biopolymers, such as polysaccharides and proteins, as well as their complexes, are amphiphilic macromolecules that combine both polar and hydrophobic fragments, have surface-active properties, low toxicity and excellent biocompatibility, thus they can be considered as promising stabilizers for food emulsions. A special place among polysaccharides is occupied by chitosans and alginates, which, in addition to other advantages mentioned above, are accessible and cheap materials.

The purpose of this work was a brief overview of the prospects for using chitosan, sodium alginate and protein-polysaccharide complexes as stabilizers for emulsions and foams for food application. The article discusses the possibility of using chitosan, sodium alginate, propylene glycol alginate, as well as various protein-polysaccharide complexes as stabilizers for heterogeneous food systems, foams and emulsions, which are the basis of many food products. In addition, special attention is paid to the prospects for the introduction of polysaccharide-based emulsifiers into industrial production and the problems that must be solved for the successful development of emulsions stabilized by biopolymers, which are the basis for the creation of food products, are discussed.

Keywords: Chitosan, Sodium Alginate, Protein-Polysaccharide Complexes, Food Emulsion, Stabilization

Funding: The work was funded by Russian Science Foundation, project No. 23-64-10020.

For citation: Lavlinskaya M. S., Sorokin A. V., Holyavka M. G., Zuev Yu. F., Artyukhov V. G. Stabilization of food emulsion by polysaccharides and protein-polysaccharide complexes: a short review. *Condensed Matter and Interphases*. 2024;26(2): 187–196. https://doi.org/10.17308/kcmf.2024.26/11933

Для цитирования: Лавлинская М. С., Сорокин А. В., Холявка М. Г., Зуев Ю. Ф., Артюхов В. Г. Стабилизация пищевых эмульсий полисахаридами и белок-полисахаридными комплексами: краткий обзор. *Конденсированные среды и межфазные границы*. 2024;26(2): 187–196. https://doi.org/10.17308/kcmf.2024.26/11933

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1. Introduction

Emulsions, which are heterogeneous disperse systems, usually consist of two immiscible liquids, where one of them, the dispersed phase, is distributed in the form of droplets in the other, the continuous phase [1]. The use of emulsion systems is widespread in the food industry, for example, to improve the flavor characteristics of products, protect and deliver biologically active substances, and they are also the basis of some food products, such as mayonnaise, sauces, and creams [2].

The main problem that arises in the preparation and practical use of emulsions is their thermodynamic instability, expressed as destabilization and phase separation [3]. The addition of emulsifiers with surfactant and/or thickening properties ensures the formation of a stable emulsion. Now, synthetic surfactants are widely used to stabilize emulsions; however, their use can have a negative effect on the organism of consumers [4]. Thus, the possible binding of anionic surfactants to proteins, enzymes, and phospholipid membranes can lead to changes in the structure of human proteins and the dysfunction of enzymes and phospholipid membranes [5]. According to cytotoxicity tests, nonionic surfactants have fewer toxic effects than cationic, anionic and amphoteric surfactants, while the toxicity of cationic surfactants is the highest [6]. Therefore, replacing synthetic surfactants with biocompatible amphiphilic compounds is a key point in expanding the scope of application of emulsions in the creation of functional foods.

The use of natural polymers such as proteins, polysaccharides, or their complexes as emulsifiers and/or stabilizers of food emulsions appears to be a promising approach to obtain highly digestible products [3]. The most common emulsifiers currently used in the food industry are a mixture of a low molecular weight surfactant, a natural amphiphilic polymer and an auxiliary co-emulsifier [7]. Polysaccharides such as pectin, various gums, and galactomannans are used as amphiphilic polymers. [8, 9]. The choice of polysaccharide as a natural component is due to the fact that, compared to protein, the stabilizer of most emulsions of natural origin, polysaccharides form a more voluminous hydration layer, leading to the increased stability of emulsions due to the structural factor [10]. In addition, the low digestibility of polysaccharides in the gastrointestinal tract slows down the rate of release of biologically active substances [11].

Thus, emulsions currently used in the food industry based on polysaccharides are multicomponent mixtures containing various synthetic low-molecular compounds. Therefore, the search for new alternative surfactants is an urgent task for the modern food industry, aimed at creating functional food products that combine not only energy value, but also biologically active additives. Promising polymers are chitosans and alginates, polysaccharides of marine origin, solutions of which have sufficient viscosity to stabilize two-component emulsion systems. At the same time, the use of protein-polysaccharide complexes allows combining the properties of both components of the system, regulating both the rheological and surface-active properties of the system and the affinity for biologically active substances, expanding the range of functional components (vitamins, antioxidants, flavonoids, etc.), implemented into the final product.

The purpose of this work was a brief overview of the prospects for using chitosan, sodium alginate, and protein-polysaccharide complexes as stabilizers for emulsions and foams for food use.

2. Use of chitosan, alginates and proteinpolysaccharide complexes as emulsifiers 2.1. Chitosan and its use as a stabilizer for food emulsions

Chitosan is a randomized copolymer of D-glucosamine and *N*-acetyl-D-glucosamine, interconnected by 1,4- β -glycosidic bonds, is a product of deacetylation of the natural polymer chitin - poly-*N*-acetyl-D-glucosamine, one of the most common natural polysaccharide, found in the shells of crustaceans and the fungus cell walls [12]. Significant scientific and practical interest in chitin and chitosan is due to their unique properties such as biocompatibility, low toxicity, biodegradability, and high sorption capacity for heavy metals and radionuclides [13].

The properties of chitosan are significantly influenced by its molecular weight (MW), which for unmodified polymers obtained from natural M. S. Lavlinskaya et al.

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chitin is in the range of 2–1000 kDa. Another equally important parameter that determines the ability of chitosan to dissolve in acidic media is the degree of deacetylation (DD). For a polysaccharide with a DD higher than 55 %, the dissolution in a 1% acetic acid solution is typical. Solubility is due to the protonation of the primary amino group at the C-2 position of the D-glucosamine unit; thus, in an acidic medium, chitosan is converted into a polycation, which is a rare phenomenon for natural polysaccharides [13].

The polymer is limitedly soluble in water; this is determined by the degree of deacetylation and the molecular weight. According to literature data, chitosan with an MW of less than 50 kDa and a DD of more than 80% is soluble at acidic and neutral pH values, and with a DD of more than 55% it is soluble at a pH below 6.5 (pK_a for chitosan) regardless of the molecular weight [13].

The diverse biological activity of chitosan, along with its safety for humans, determines the widespread use of this polysaccharide in the food industry. The antioxidant and antimicrobial activity of chitosan, as well as its ability to interact with various compounds [14-16], allow it to be used for the development of "smart packaging" that increases the shelf life of food products; its high emulsifying ability allows to replace synthetic surfactants in food technologies. The earliest mention of the use of chitosan in the food industry in patent data dates back to 1956: in a patent, chitosan hydrochloride was used as a modifying agent in the preparation and creation of chewing gum [13].

Chitosan is an effective emulsifier for stabilizing heterogeneous oil-in-water systems. The polysaccharide increases the viscosity of the dispersed phase, complicating the diffusion of dispersed particles and reducing the rate of droplet aggregation. In addition, the positively charged amino groups make chitosan an amphiphilic surfactant polymer. Chitosan can be used as the only emulsifier; however, the resulting emulsions are reversible due to sensitivity to pH. In an acidic media with pH < pK_a protonated chitosan forms polyelectrolyte complexes (PEC), interacting with the carbonyl groups of triglycerides, and in the case of increasing pH to values above pK_{a} PEC destruction and loss of emulsifying ability occurs. With increasing pH, the solubility of chitosan decreases, and the emulsion remains stable due to the oil phase droplets adsorbed on the chitosan particles. With a reverse increase in the acidity of the medium, chitosan transforms into a soluble form, desorbing dispersed oil droplets, and the emulsion reversibly stratifies [17]. The emulsifying ability of chitosan largely depends on the degree of deacetylation and molecular weight: it increases for low-molecular chitosans at DD of less than 60% and more than 86%, while at DD values from 65 to 77% these properties significantly depend on the concentration of the polysaccharide [18]. In a majority of experimental studies on the use of chitosan as an emulsifier, it was considered as a component of complex compositions containing other surfactants. The presence of both chitosan and protein emulsifiers makes the heterogeneous system more stable. Using a mixture of chitosan with soy protein isolate can improve the digestibility and stability of emulsified carotenoids [19]. A complex containing chitosan modified with β -lactoglobulin fibers stabilizes fish oil emulsion in water [20]. It has also been shown that stable Pickering emulsions, emulsions in which solid particles containing corn oil act as a stabilizer, are formed when the polyelectrolyte complex of chitosan and gelatin is used as an emulsifier [21]. The ability of chitosan to form polyelectrolyte complexes in aqueous solutions can be used to increase the stability of easily degradable compounds, such as carotenoids [22] and anthocyanins [23].

2.2. Alginates and their use for stabilization of food emulsions

Sodium alginate, the sodium salt of alginic acid, is a recognized food ingredient widely used in the production of functional foods. As a food ingredient, the use of alginate is based on three main properties: the ability to form thick solutions, gelation, and film formation. The considered polysaccharide is widely used for the production of many new functional food products, such as food jelly, restructured meat, packaging and protective materials for packaged, sliced, or prepared fruits, vegetables, etc. In addition, new applications of sodium alginate may arise after its chemical, physical and biological modifications, leading to the production of derivatives with the required functional properties. M. S. Lavlinskaya et al.

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Sodium alginate is a polysaccharide isolated from brown algae, where it occurs as a component of the cell wall, performing structural functions similar to carrageenans and agar [24]

Being a polymeric acid, alginate consists of 1,4-linked α -L-guluronic acid (G-units) and β -D-mannuronic acid (M-units) residues. These two acidic residues differ greatly in stereochemistry at the C-5 atom. Alginates obtained from different algae species differ in the content of G- and M-units, present in the polymer chain in the form of GG, MM and MG/GM blocks in different ratios, which leads to differences in the physical properties of alginate gels [25] and the characteristics of alginate-based products [24].

The wide practical use of alginate is based on its three main properties. Firstly, it is the ability to increase the viscosity of aqueous solutions. The second is the ability to turn into a gel when salts of divalent cations of various metals are added [25, 26] to an aqueous solution of sodium alginate. Unlike carrageenan or agar gels, temperature changes are not required for the formation of a heat-stable alginate gel, which saves not only energy, but also protects biologically active substances from thermal destruction. The third practically significant property of sodium alginate is the ability to form films and fibers. In addition, the unique structure of this polymer is biocompatible [27].

As a natural water-soluble polymer, alginate forms viscous aqueous solutions. The thickening properties of alginate are commonly used in the production of jams, marmalades and fruit sauces since the interactions between alginate and pectin are reversible upon heating and provide a higher viscosity than either component alone. Alginates are also used to thicken desserts and sauces such as mayonnaise. The use of alginate alone or in combination with other thickeners improves the organoleptic characteristics of a number of low-fat foods. The hydrophilic nature of alginate helps to retain water and improves the texture of food, resulting in improved food acceptance by consumers [28].

As a gelling agent, alginate forms stable gels over a wide temperature range and at low pH values, which can be used in the food industry. The introduction of alginates into the formulation of culinary creams provides resistance to freezing/thawing and reduces the separation of solid and liquid components. In ice cream, alginate is often used in combination with other hydrocolloids for thickening and stabilization, which allows controlling the viscosity of the product, increasing resistance to heat shock, and reducing shrinkage and ice crystal formation. In addition, alginate is widely used in the creation of artificial products, for example, an analogue of fish caviar [28].

As an emulsifier in industry, it is currently not being proposed to use alginic acid or its salt, but a chemical modification product, propylene glycol alginate (PGA) (Fig. 1). This compound is an esterified alginate derivative, widely used in the food and beverage industry. PGA was first obtained by Kelco in 1949 [29].

Since in this compound the carboxyl group of alginic acid is substituted by propylene glycol ether, PGA dissolves in an acidic medium up to pH 3-4, under conditions under which sodium alginate precipitates as alginic acid. Resistance to acidic conditions and to the ionic strength of solution makes propylene glycol alginate a valuable component in foods and beverages with high acidity or divalent metal ion content. In addition, PGA also has high lipophilicity and emulsifying ability due to the propylene glycol moiety contained in its molecules.

The addition of 0.1% propylene glycol alginate increases the colloidal stability of fruit and vegetable juices without compromising their taste

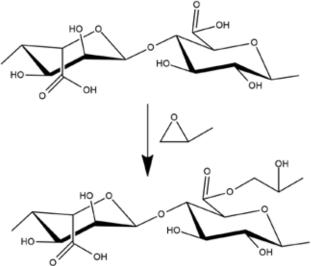


Fig. 1. Scheme of the propylene glycol alginate synthesis

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and composition. Due to the high emulsifying characteristics of propylene glycol alginate, it is possible to obtain concentrates and juices enriched with dry substances (pulp, etc.), both hydrophilic and lipophilic, which has a beneficial effect on the consumer characteristics of the final product [28].

In the literature there is also information about the use of propylene glycol alginate in a mixture with some other polysaccharides (carboxymethyl cellulose, gums, etc.) as a stabilizer for protein drinks, beer foam, and soy milk [28]. It is noted that the required amount of stabilizer does not exceed 0.5%, out of which more than 60% is propylene glycol alginate, which does not affect the taste and consistency of the final product.

In addition, propylene glycol alginate has proven itself as a stabilizer for yoghurt with fruit fillings. Due to the acidic pH, the choice of stabilizers is quite limited, but in addition to maintaining high system uniformity and resistance to separation, the emulsifier provides a marketable appearance to the product [28].

The presence of hydrophobic fragments in the macromolecules of propylene glycol alginate, the latter acts as an effective stabilizer for salad dressings, improves the external and organoleptic characteristics of bread and the texture of pasta, reducing the pasta fragility [28, 30].

2.3. Stabilization of food emulsions and foams with protein-polysaccharide complexes

Protein-polysaccharide complexes combine the physicochemical and functional properties of their constituent macromolecules and, accordingly, hydrophobic and hydrophilic properties. Consequently, they can be successfully used as agents stabilizing the air/water or oil/ water interfaces in complex food systems [31].

Schmitt et al. [32] studied the surfactant properties at the air/water interface of β -lactoglobulin/acacia gum complexes prepared at pH 4.2 and a component ratio of 2:1 and compared them with the behavior of β -lactoglobulin. The surface activity of the complexes was similar for the value corresponding to the protein; however, the complexes formed stronger viscoelastic films with a thickness of about 250 Å at the interface. As a result, the gas permeability of films obtained

from associates was lower compared to films from native β -lactoglobulin. In addition, by reducing the rate of aggregation of air bubbles, the complexes stabilized water-air foams. The results obtained were used to formulate complex food products, such as fruit ices and sorbets, for which the stability of air bubbles in the foam correlates with improved organoleptic parameters of the product. Complexes of whey protein isolate and acacia gum exhibit similar properties, which allows to use them for the replacement of animal gelatin when creating vegetarian foods [33].

Complexes based on β -lactoglobulin and pectins are also used to stabilize the air/water interface [34]. In this case, the charge density of pectin, i.e. the degree of its methylation and the ratio of components determine the size of the resulting complexes and, consequently, their surface activity. The viscoelastic properties of the air/water interface are determined either by the adsorption of the complex or by the sequential adsorption of the components. In the latter case, the formation of viscoelastic films was observed. A study of the structure of the resulting films showed that both obtained samples contain a dense layer near the air/water interface, which probably consists of β-lactoglobulin. However, the thickness of the film obtained with sequentially adsorbed components is higher than for adsorbed complexes. It should be noted that associates based on ovalbumin and pectin or β-lactoglobulin and carboxylated pullulan also exhibit surfaceactive properties at the water-air interface and can be used to stabilize foam. It has been shown that napin, a protein isolated from rapeseed flour, forms complexes with pectins, which also stabilize aqueous foams and have a higher surface activity compared to the native protein [35].

Emulsions, widely used in the food industry, can also be stabilized with protein-polysaccharide complexes. If the complex is formed during the emulsification step, a mixed emulsion is produced [31]. There is also a layer-by-layer emulsion stabilization technique: in this case, the primary emulsion is stabilized by protein, and then a polysaccharide dispersion is added, inducing interfacial complexation, leading to the formation of so-called bilayer emulsions [36]. This approach is most commonly used on an industrial scale because it produces stable emulsions for a wide M. S. Lavlinskaya et al.

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range of compounds. A study of the rheological behavior of the sodium caseinate-dextran sulphate complex showed that the resulting interfacial layers at the interface are much more viscoelastic in the case of mixed emulsions [37]. Interestingly, the resulting emulsions showed different pH stability, especially in an acidic medium: mixed emulsions turned out to be much more resistant to flocculation compared to double-layer emulsions. These results have practical implications for the in vivo control of lipolysis. Ducel et al [38] reported high surface activity in an oil/water system for complexes of pea globulin or α -gliadin with acacia gum. The resulting films were characterized by a long relaxation time and high elasticity. In addition, complexes obtained at a lower pH stabilize emulsions more effectively due to having a higher spreadability on the surface of oil droplets.

Cho and McClements [39] emphasized the importance of controlling the ratio of protein to polysaccharide and the concentration of the latter to ensure the colloidal stability of a bilayer emulsion obtained in the presence of a β-lactoglobulin-pectin complex at pH 3.5. Too low (<0.02%) or high (>0.1%) pectin content led to the formation of an unstable emulsion due to flocculation. Another study showed that the presence of 100 mM NaCl and β -lactoglobulin/ citrus pectin complex produced more stable emulsions at pH 3-4 compared to emulsions stabilized by β -lactoglobulin alone. This is explained by the fact that in the presence of an electrolyte, shielding of the interfacial charge is achieved [40]. Some other protein-polysaccharide complexes (*β*-lactoglobulin with alginate, ι-carrageenan or acacia gum) have been used to obtain acid-resistant bilayer emulsions and for industrial beverage production [41, 42].

The use of polycationic chitosan allows obtaining stable emulsions based on whey protein isolate at pH 6.0 [43]. As with other described systems, the ratio of protein and polysaccharide in the complex used plays an important role in stabilization. Interestingly, such systems form at a much lower pH (around 3.0) and can reduce surface tension as well as pure proteins. Stable concentrated emulsions containing up to 40% rapeseed oil can be obtained using a wide range of biopolymer concentrations ranging from 3.8 to 11.2%, which allows additional control of the volumetric viscosity of the resulting product. The stability of an emulsion containing 15% sunflower oil, stabilized with whey protein isolate, increases when using a complex of chitosan and acacia gum at pH 3.0. The use of the complex leads to the formation of monodisperse droplets, gel-like emulsions or clusters of oil droplets, depending on the ratio of chitosan to acacia gum [44].

A study of the emulsifying properties of a complex of whey protein isolate and carboxymethyl cellulose revealed a dependence on the protein/ polysaccharide ratio for heterogeneous systems containing 10 and 20% oil fraction. It should be noted that the use of these complexes allows obtaining heat-stable emulsions, which is not observed for the stabilization with pure protein [45].

Complexes of soy protein isolate and hydrophobically modified pectin stabilize emulsions at pH 5.5. It has also been shown that protein-polysaccharide complexes are capable of stabilizing the outer interface of W/M/W emulsions obtained at pH \leq 6.0 [45].

3. Problems and prospects for using polysaccharides for the stabilization of food emulsions

The complex compositions of food systems determine the high demands placed on emulsifiers, among which health and the environment safety, as well as the ability to maintain emulsifying properties during food processing, are especially important. Researchers proposing polysaccharides for the stabilization of emulsions are faced with problems of their interaction with other components of systems (electrostatic interactions with salts and proteins, the formation of hydrogen bonds with other macromolecules, hydrophobic interaction with polyphenols, etc.), and they are also subject to thermal destruction as a result of processing food products. Despite the fact that the principles of the influence of external factors on the emulsifying properties of polysaccharides seem to have already been finally established, the significantly different molecular structure of polysaccharides leads to significant variations in their emulsifying properties. Therefore, in the case of using a new, previously undescribed polysaccharide

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as a food emulsifier, a full investigation of its colloidal chemical characteristics is required. In addition, there is virtually no information in the literature on changes in the emulsifying properties of polysaccharides during food processing (thermal or non-thermal), as well as on the molecular mechanisms of these changes. Thus, the introduction of natural polysaccharides as food emulsifiers is undoubtedly a laborintensive task.

Currently, emulsion systems obtained using polysaccharides as emulsifiers are used mainly to create functional foods capable of targeted delivery and the stabilization of active substances, as well as to study the mechanisms of their action, release, digestion, absorption, and transportation. Therefore, the development of a functional product should also include many stages of research, such as determining the structural characteristics and interfacial behavior of the polysaccharide, emulsion characteristics (stability, droplet size, etc.), as well as studying the kinetics of metabolism and the bioavailability of released biologically active substances. The result of solving these problems can be used to create emulsion systems that could specifically deliver the optimal amount of biologically active substances, protecting them from destruction during movement through the gastrointestinal tract [31].

4. Conclusion

Thus, polysaccharides, in particular chitosan and alginic acid derivatives, as well as proteinpolysaccharide complexes, are promising for the development of multifunctional emulsion systems capable of not only maintaining the stability of a heterogeneous system for a long time, but also giving them functional properties, for example, as nano- and microcontainers for biologically active substances. The introduction of polysaccharidebased stabilizers into the production of food is difficult due to the labor-intensive processes for creating such systems. However, the fact that sodium alginate and chitosan are already successfully used in food technology as thickeners and components for "smart" and environmentally friendly food packaging increases the potential for these polysaccharides to find early use in other sectors of the food industry.

Contribution of the authors

The authors contributed equally to this article.

Conflict of interests

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

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Received 13.09.2023; approved after reviewing 06.11.2023; accepted for publication 15.11.2023; published online 25.06.2024.

Translated by Valentina Mittova