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Complexes of Porcine Gelatin with Carrageenans: Influence of pH, Presence of Salts, and Polysaccharide Type

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Abstract–The effect of the polysaccharide:protein mass ratio, pH, concentration, and nature of the inorganic salt (sodium and calcium chlorides) on the phase behavior of aqueous mixtures of porcine gelatin with kappa-, lambda-, or iota-carrageenans was studied using turbidimetric titration. The formation of insoluble protein-polysaccharide complexes was observed in the range of pH between two boundary values, which decreased with increasing polysaccharide–protein mass ratio (Z). An increase in the number of sulfate groups in the polysaccharide in the series kappa- < iota- < lambda-carrageenan leads to a decrease in the boundary pH values at constant Z. Electrostatic interactions between gelatin and carrageenan molecules are suppressed with increasing ionic strength, which leads to the destruction of insoluble complexes upon addition of salts.

Keywords: porcine gelatin, carrageenans, polyelectrolyte complexes, turbidimetric titration, protein-polysaccharide interactions

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INTRODUCTION

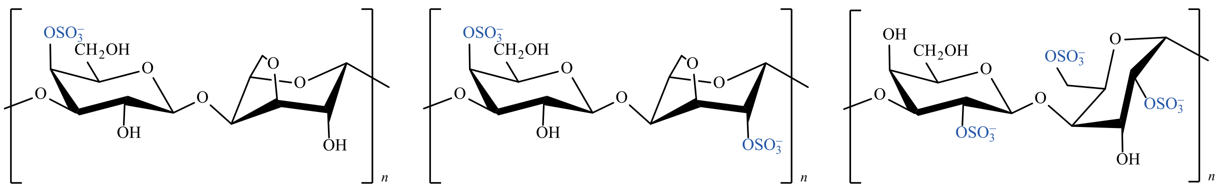
With the increasing demand for healthy nutrition, particularly with a reduced fat, sugar, and salt content, the food industry has to deal with the challenge of developing products that could satisfy consumer interests while maintaining good organoleptic properties and high nutritional value [1–4].

The use of protein-polysaccharide complexes, which offer a number of advantages, represents a promising solution to this problem. Complexes of these biomacromolecules perform structural functions in food products, which allows to create improved product textures [5, 6]. The combined use of protein with polysaccharides alters the rheological properties [7–9], which is especially important for food emulsions and gels. Protein-polysaccharide complexes retain water effectively, which helps to develop low-fat products [10, 11] and enhance the stability of the final product [12–14], especially under changing storage and transportation conditions.

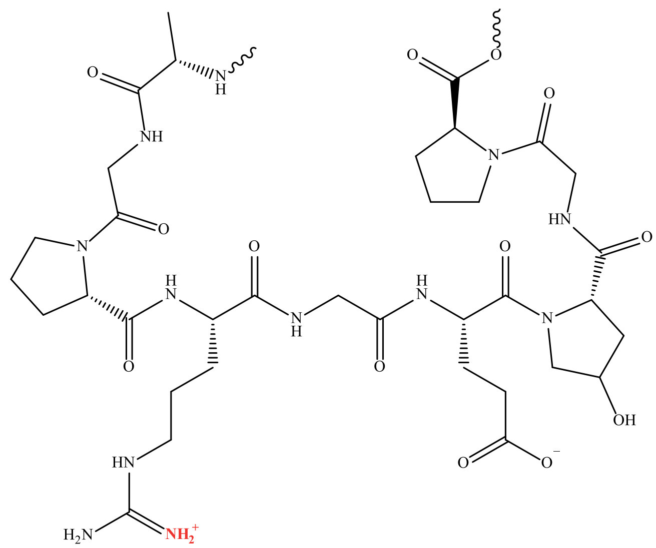
It is important to understand the influence of various factors on the formation and functional properties of protein-polysaccharide complexes. Examples of such factors include mixture composition, pH value, salt concentration, mixing temperature, and the presence of different additives [7, 15–18]. Therefore, studies examining the influence of these factors on the structure and properties of the resulting complexes are helpful to select the optimal conditions for their preparation in the food industry and other biotechnological fields.

In this paper, we analyze the effects of mass ratio, pH, inorganic salt concentration, and type of carrageenan on the complexation of carrageenans with porcine gelatin. Porcine gelatin has a neutral taste and odor, and is more affordable than beef and fish gelatin, which makes it a common ingredient in the food industry [19]. Three types of carrageenans are considered: iota (ι), lambda (λ), and kappa (κ). Both carrageenans and gelatin have irregular structure. The structures of their representative fragments are shown in Scheme 1. The primary difference between the carrageenans is the number of sulfate groups per two galactose residues: λ-carrageenan has on average three

Scheme 1.



κ-Carrageenan ι-Carrageenan λ-Carrageenan



Ala-Gly-Pro-Arg-Gly-Glu-4Hyp-Gly-Pro

sulfate groups, ι-carrageenan has two, and κ-carrageenan has a single group. This directly affects their properties, such as dissolution temperature, gelation ability [20], and intermolecular interactions with proteins [21]. Previously, the influence of the type of carrageenan on the formation of protein-polysaccharide complexes has only been studied for canola protein isolate [22]. For porcine gelatin, only the complex with κ-carrageenan was studied [9, 23].

RESULTS AND DISCUSSION

Influence of the polysaccharide–protein mass ratio. During turbidimetric titration of porcine gelatin with carrageenans, the dependence of optical density (Fig. 1a) and pH (Fig. 1b) on the mass ratio of carrageenan to porcine gelatin (Z) was determined.

At the beginning of the titration, an increase in optical density was observed for all carrageenans, which is due to an increase in the amount of insoluble protein-polysaccharide complexes. During the formation of these complexes, positively charged amino groups of gelatin, primarily arginine [24–26] (Scheme 1), are neutralized by the negatively charged sulfate groups of carrageenan.

The maximum optical density value for each type of carrageenan is observed at different polysaccharide:protein mass ratios: Z = 0.04 for λ-carrageenan, Z = 0.06 for ι-carrageenan, and Z = 0.2 for κ-carrageenan. If we consider the dependence of pH on the polysaccharide:protein mass ratio (Fig. 1b), it can be seen that the maximum optical density is achieved at approximately the same pH value of 5.6 for different carrageenans, which corresponds to the isoelectric point of porcine gelatin. This appears to be an interesting feature of the studied systems. The amount of carrageenan required to form a polyelectrolyte complex (PEC) depends on the charge of its anion determined by the number of sulfate groups. For λ-carrageenan, which contains the largest number of negatively charged groups (Scheme 1), the mass ratio of polysaccharide to protein for the formation of PEC is the smallest in the studied series of carrageenans, and for κ-carrageenan with a single sulfate group per two saccharide units, it is the largest. The value of optical density at the point of PEC formation is also the highest for the complex of κ-carrageenan with

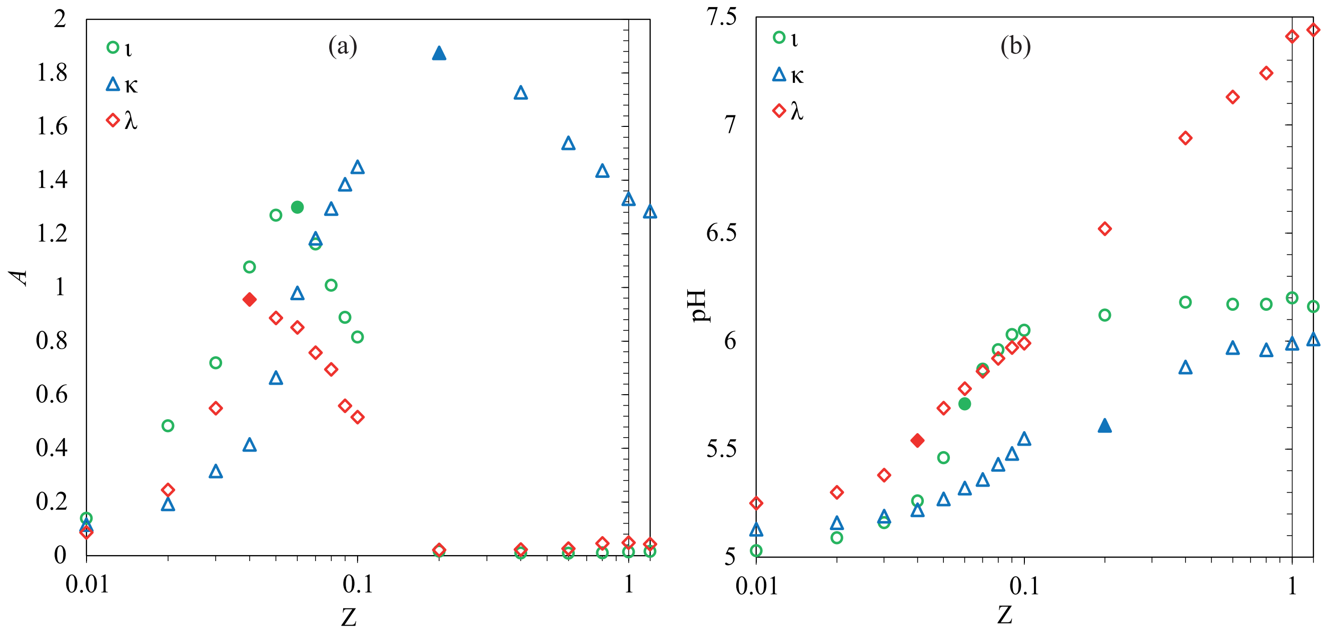


Fig. 1. Dependence of (a) optical density (t = 25°C, λ = 400 nm), (b) pH value in aqueous solutions of porcine gelatin (0.1% w/v) upon addition of carrageenan solution on carrageenan–gelatin mass ratio (Z). ι-carrageenan—green circles, κ-carrageenan—blue triangles, λ-carrageenan—red diamonds.

porcine gelatin, and the lowest for λ-carrageenan. This can be explained by a larger mass of precipitate formed at higher Z values.

With the further addition of carrageenans, the pH value becomes higher than the isoelectric point of gelatin, and its overall charge becomes negative, which leads to the repulsion of gelatin molecules from carrageenan molecules. At such conditions, the insoluble complexes become less stable and the optical density starts to decrease. At high polysaccharide:protein mass ratio ($Z >$ 0.2), λ- and ι-carrageenans do not interact with gelatin, as evidenced by the optical density close to zero (Fig. 1a).

Previously, Kolotova et al. [16] used turbidimetric titration to find that the maximum optical density of a mixture of fish gelatin and sodium alginate is observed at a mass ratio of polysaccharide to protein equal to 0.07. In a study of the structure and rheological properties of κ-carrageenan complexes with fish gelatin [8], the highest optical density was observed at a protein to polysaccharide ratio of 96 : 4 (Z = 0.04). The zeta potential of this mixture was close to neutral indicating predominantly electrostatic interactions during complex formation. A study of the complex formation of fish gelatin with hyaluronic acid by Razzak et al. [27] showed that the contribution of non-electrostatic interactions, namely hydrophobic interactions and hydrogen bonds, is insignificant. In the FTIR spectra of the κ-carrageenan-gelatin mixture, a slight shift of the protein bands along with the polysaccharide bands is observed, indicating chain-chain interactions mediated by electrostatic interactions [18]. It is interesting to note that gelatin forms soluble complexes above the protein isoelectric point with such anionic polysaccharides as sodium alginate [27], hyaluronic acid [28] and gum arabic [29]. This is also possible in the case of carrageenans.

Influence of pH. As noted in the previous section, the pH of the solution strongly influences the complexation between gelatin and polysaccharides. The pH value determines the charges of both molecules and, therefore, their ability to form stable electrostatic complexes.

We studied the effect of pH on the formation of protein-polysaccharide complexes for each type of carrageenan by adding an acid or alkali solution to a solution of the mixture of components at the fixed carrageenan:gelatin mass ratio (Fig. 2). Insoluble polyelectrolyte complexes of protein with polysaccharides are formed in the pH range between the isoelectric point of the protein (≈5.6) and the polysaccharide pK_a value (1–2). Such conditions promote the attraction of oppositely charged macromolecules to each other. Dissolution of the complexes at lower pH

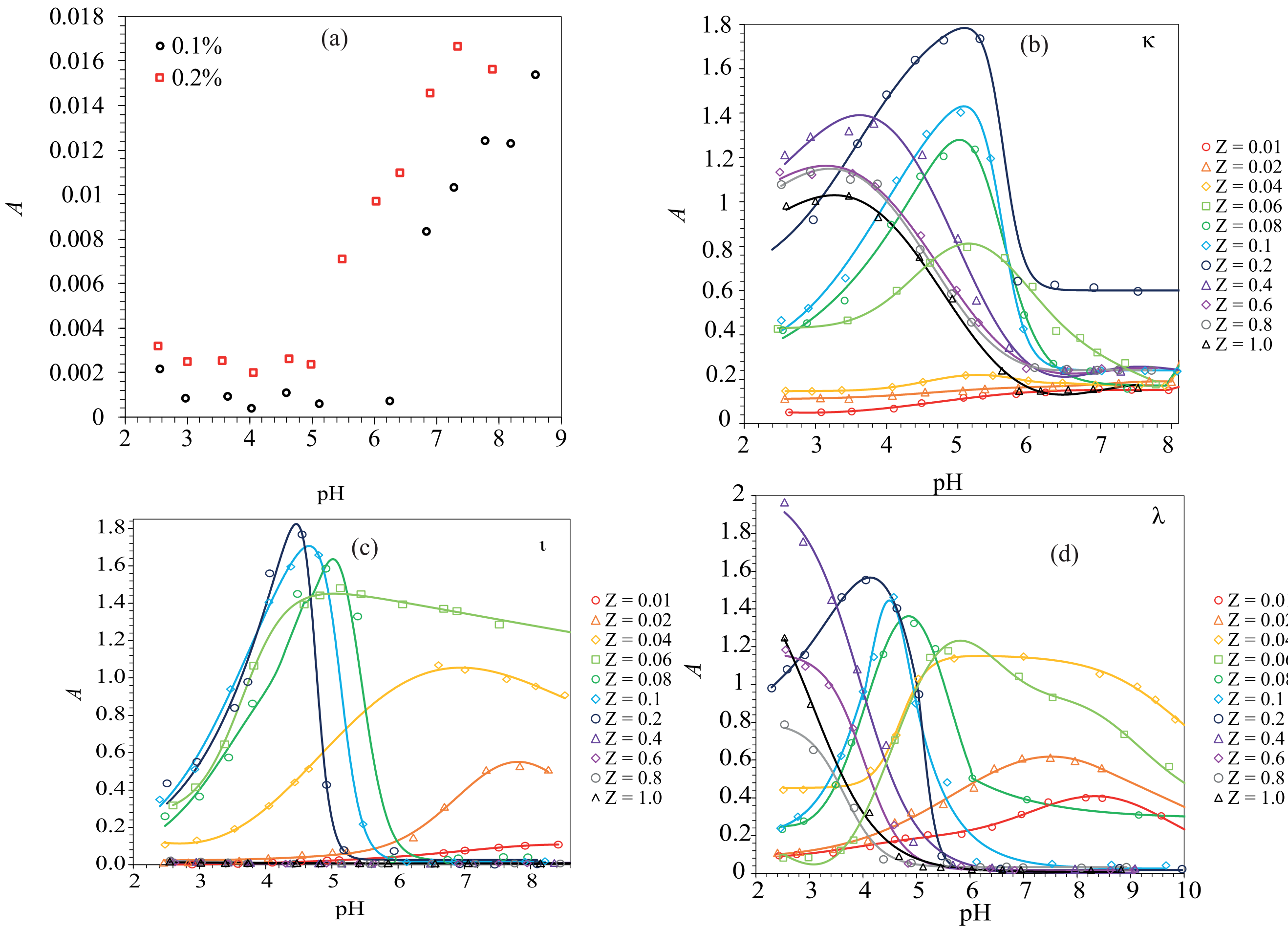


Fig. 2. Changes in the turbidity of gelatin solutions with varying pH (a) and dependence of the optical density on the pH value in aqueous solutions of porcine gelatin (0.1% w/v) in the presence of carrageenans at different carrageenan–gelatin mass ratio (Z) for κ-carrageenan (b), ι-carrageenan (c), λ-carrageenan (d) at 25°C (λ = 400 nm).

values can be caused not only by further protonation of the polysaccharide but also by an increase in the ionic strength of the solution. It should be noted that the turbidity of individual porcine gelatin solutions remains essentially unchanged with varying pH (Fig. 2a).

With an increase of Z, the peaks in the curve of the dependence of optical density on pH corresponding to the formation of insoluble protein-polysaccharide complexes, shift to the acidic pH values. The boundary and the corresponding maximum optical density pH values ($pH_{\varphi 1}$, $pH_{\varphi 2}$, pH_{opt}) decrease. The dependence of the boundary pH values on the mass ratio Z (Fig. 3) is exponential. It is worth noting that the magnitude of the shift of the boundary pH values depends on the number of sulfate groups: the complex of λ-carrageenan with gelatin undergoes the largest shift, and the complex with κ-carrageenan shifts less. For ι-carrageenan–gelatin complexes with a mass ratio $Z > 0.2$, the optical density is close to zero over the studied range of pH values. It was previously noted that ι-carrageenan has the greatest effect on inhibiting protein complexation among the three carrageenans under consideration [22, 30], and the magnitude of electrostatic interactions between the three types of carrageenans and the protein depends not only on the relative charge density but also on the conformation of the polysaccharide [31]. A decrease in the boundary pH values with increasing polysaccharide concentration was also described by Razzak et al. for a mixture of fish gelatin with sodium alginate [27], as well as in a study of the binding of canola protein to κ-, ι-, and λ-carrageenans [22]. Zeta potential measurements by Gonçalves de Alcântara et al. [32] also indicate an electrostatic pattern: the gelatin-κ-carrageenan hydrogel formed below the isoelectric point of the protein. This effect is linked to

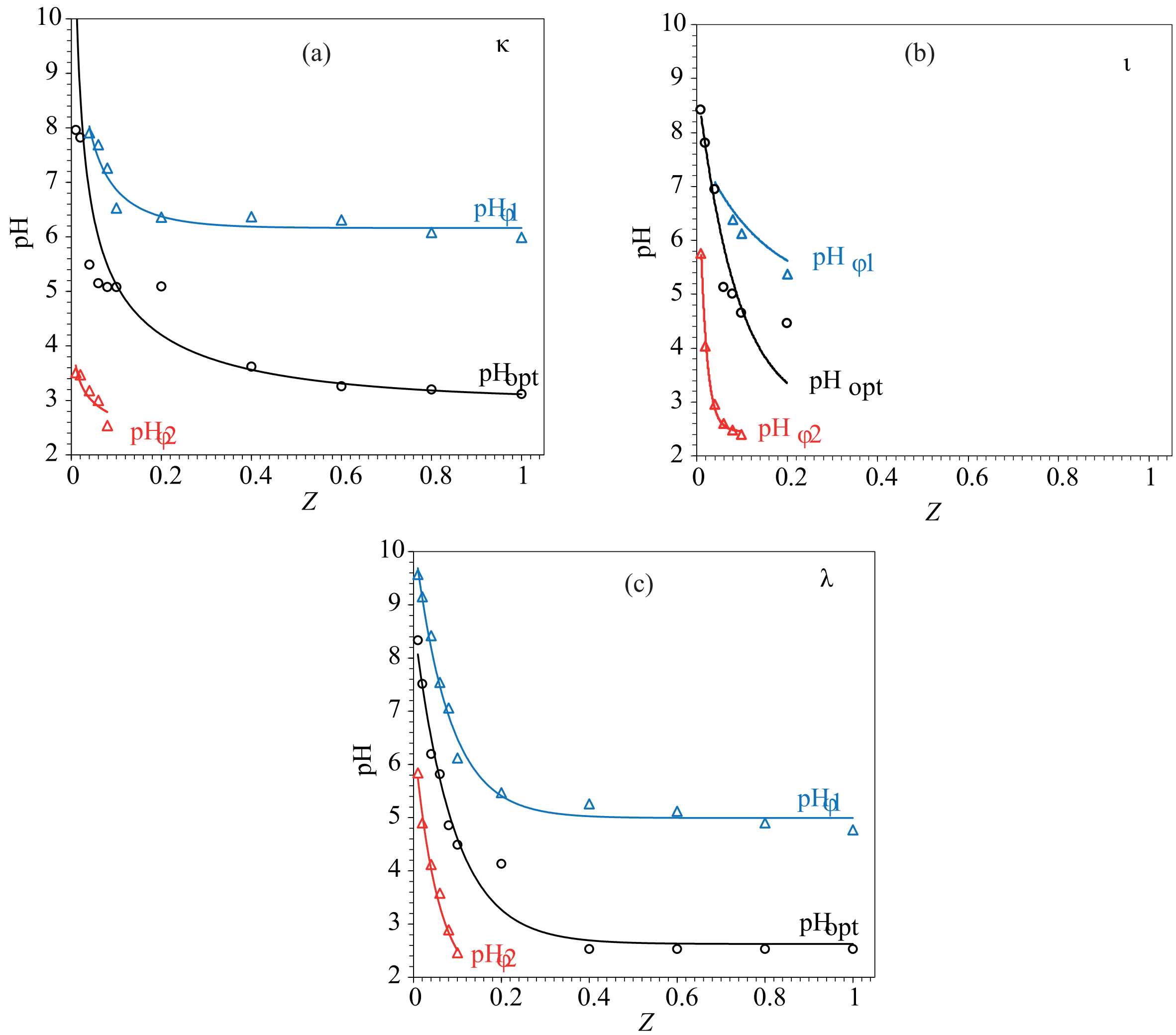


Fig. 3. Changes in the boundary pH values for complex formation in aqueous mixtures of porcine gelatin (0.1% w/v) at different carrageenan–gelatin mass ratio (*Z*) for κ-carrageenan (a), ι-carrageenan (b), and λ-carrageenan (c).

the fact that with a decrease in pH, the positive charge of protein molecules increases, and an increasing amount of polysaccharide is required for its neutralization.

Influence of salt concentration and cation type. The presence of inorganic salts increases the ionic strength of the solution and usually decreases the stability of polyelectrolyte complexes. The effects of sodium [7, 27, 33] and calcium [16] chlorides on protein-polysaccharide mixtures are often considered since they are widely used in the food industry [35–37].

The influence of salts on the behavior of the mixtures of porcine gelatin and carrageenans with the maximum optical density was examined. The corresponding values of *Z* were 0.04 for λ-carrageenan, 0.06 for ι-carrageenan, and 0.2 for κ-carrageenan. The study was repeated with a number of concentrations of NaCl (Fig. 4) and $CaCl_2$ (Fig. 5).

With increasing salt concentration, the optical density of the mixtures decreases. The higher ionic strength of the solution significantly weakens the electrostatic interactions between the protein and the polysaccharide due to charge shielding, which leads to the dissolution of insoluble protein-polysaccharide complexes. $CaCl_2$ stronger suppresses the interactions between porcine gelatin and carrageenans due to the larger charge and radius of the cation. It is interesting to note that with an increase in ionic strength, there is also a shift in the boundary pH values of complex formation to a more

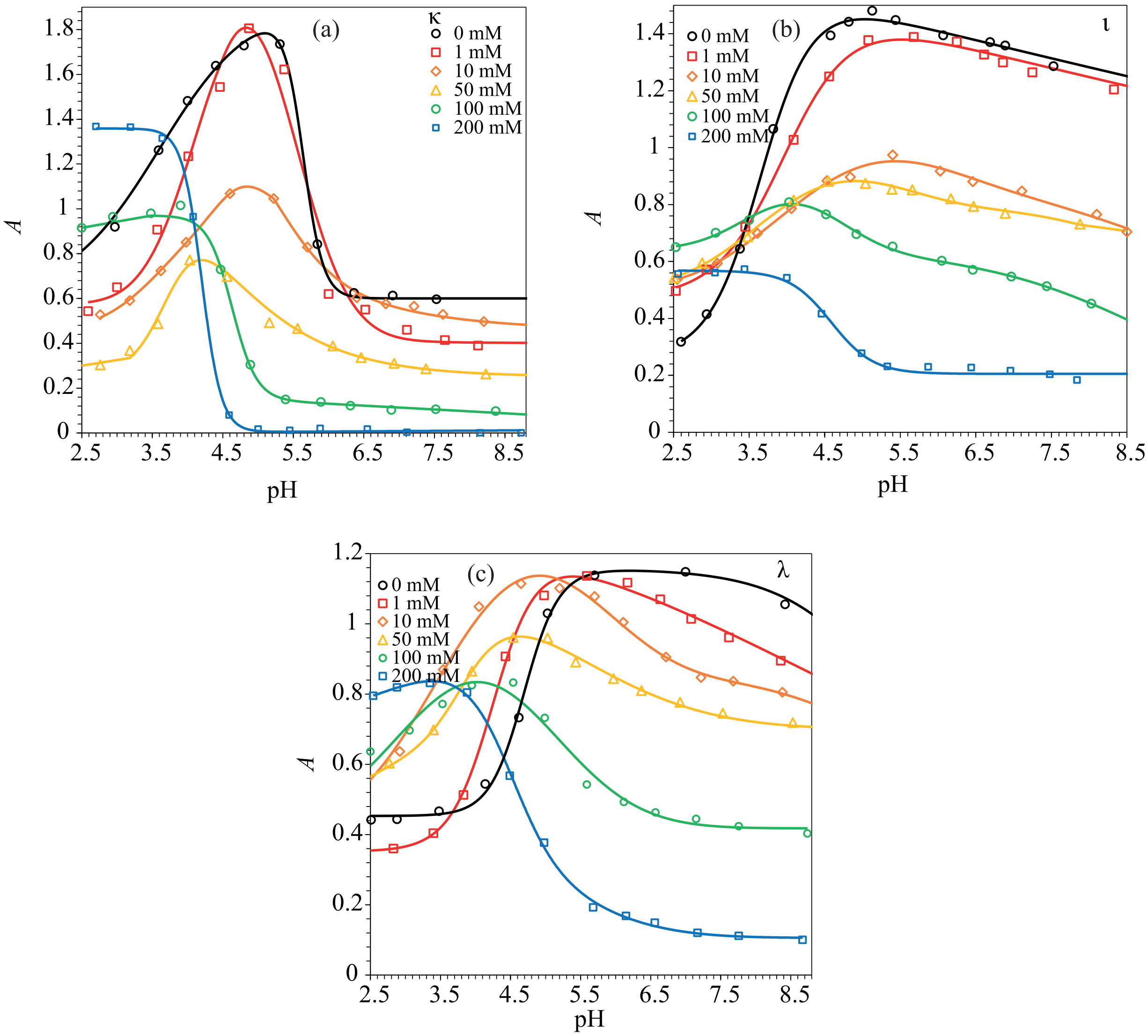


Fig. 4. Dependence of the optical density of the aqueous mixtures of porcine gelatin (0.1% w/v) on the pH value in the presence of different concentrations of NaCl and (a) κ-carrageenan (Z = 0.2), (b) ι-carrageenan (Z = 0.06), and (c) λ-carrageenan (Z = 0.04) at 25°C (λ = 400 nm).

acidic region, which is most likely associated with a decrease in the pK_a values of residues capable of protonation.

CONCLUSIONS

The study of the mixtures of carrageenans with porcine gelatin emphasizes the importance of even small changes in environmental conditions for the formation of insoluble protein-polysaccharide complexes. The results show that the type of carrageenan, the mass ratio of the components, the pH of the solution, and the concentration of salts significantly affect the interactions between the protein and the polysaccharide. The formation of complexes of porcine gelatin with carrageenans is primarily due to electrostatic interactions. Insoluble complexes are formed at pH values below the isoelectric point of the protein. The maximum amount of protein-polysaccharide complexes that can be formed corresponds to complete neutralization of the opposite charges of the biomacromolecules. The number of sulfate groups in the polysaccharide affects the quantity of polysaccharide required to neutralize the positively charged groups of porcine gelatin and the

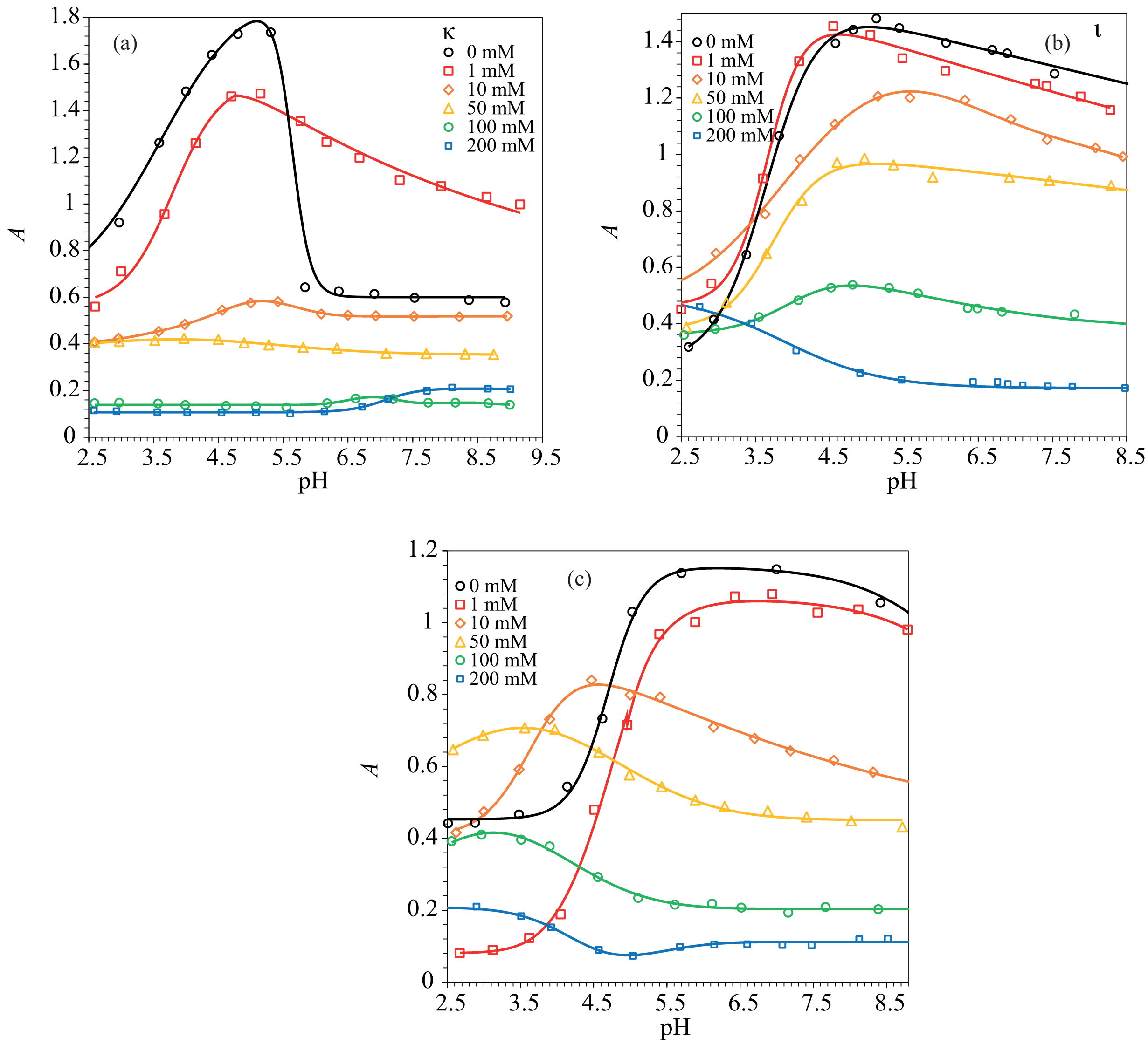


Fig. 5. Dependence of the optical density of the aqueous mixtures of porcine gelatin (0.1% w/v) on the pH value in the presence of different concentrations of $CaCl_2$ and (a) κ-carrageenan (Z = 0.2), (b) ι-carrageenan (Z = 0.06), and (c) λ-carrageenan (Z = 0.04) at 25°C (λ = 400 nm).

boundary pH values of insoluble complex formation. As the mass ratio of carrageenan to porcine gelatin increases, the boundary pH values decrease. An increase in ionic strength leads to a decrease in the yield of the polyelectrolyte complex and a decrease in the boundary pH values. This study may help to select the optimal conditions for obtaining complexes between gelatin and carrageenans. Data on the influence of the ratio of biomacromolecules, pH, and salt concentration can be used in the development of new products with improved functional properties, as well as for the optimization of production processes in the food, pharmaceutical, and biomedical industries.

EXPERIMENTAL

Porcine gelatin type A with Bloom hardness 300 (Sigma-Algrich, USA), ι-carrageenan (Alfa Aesar, USA), κ-carrageenan type I (Sigma-Algrich, USA), λ-carrageenan (TCI Europe N.V., Belgium), glacial acetic acid (≥99.5%), sodium chloride (≥99%), calcium chloride (≥98%) produced by Tatkhimprodukt (Russia) were used without prior purification.

Preparation of mixtures of porcine gelatin with carrageenans. Samples of porcine gelatin and carrageenans are left overnight to swell in deionized water at room temperature. The resulting mixtures of porcine gelatin and λ-carrageenan are thermostated at 50°C, and mixtures of ι- and κ-carrageenan are thermostated at 70°C for 1 h. The pH value of 0.2% pork gelatin stock solution is 5.02; 1% ι-carrageenan—9.57; 1% κ-carrageenan—9.65; 1% λ-carrageenan—9.51. When preparing an aqueous solution of porcine gelatin (0.1% w/v) with a polysaccharide, the initial solutions are mixed at the specified carrageenan–gelatin mass ratios (Z = 0.01–1.00) and thermostated at 40°C for 1 h.

Turbidimetric titration. The UV-vis spectra of aqueous mixtures of porcine gelatin in the absence and presence of carrageenans were recorded in the wavelength range from 200 to 800 nm at 25°C in a quartz cuvette with an optical path length of 1 cm using a Cary 100 UV-visible spectrophotometer (Agilent Technologies, USA). Deionized water was used as a reference sample. The gelatin solution (0.1% w/v) was titrated with a solution of each type of carrageenan (0.1 or 1% w/v) with constant stirring, measuring the turbidity for each resulting polysaccharide–protein mass ratio in the range 0 to 1.1. Simultaneously, the pH of the resulting solutions was measured using a HANNA HI2211-02 microprocessor pH meter (Hanna Instruments, USA).

pH-Turbidimetric titration. Aqueous mixtures of porcine gelatin in the presence of carrageenans with a mass ratio Z = 0.01 to 1.2 were titrated to pH ~ 2.5 with acetic acid solutions and to pH ~ 9 with sodium hydroxide solutions. During the titration, pH values and UV-vis spectra were recorded in the wavelength range 200 to 800 nm. Also, during titration, when adjusting the pH value and during the recording of spectra, continuous stirring was carried out, which did not allow the resulting insoluble complexes to settle at the bottom of the cuvette. The optical density and pH of the solutions were determined during titration at a wavelength λ = 400 nm and an optical path length l = 1 cm at 25°C.

The determination of the boundary pH values was carried out according to the literature [16, 34]: $pH_{\varphi 1}$ is the boundary pH value below which the mixture becomes turbid due to the formation of insoluble polysaccharide-protein complexes; $pH_{\varphi 2}$ is the boundary pH value below which the turbidity of the solution disappears due to the dissociation of these complexes; pH_{opt} is the pH value at which the highest optical density is observed.

To study the effect of the concentration and the type of inorganic salt cation on the phase behavior of the mixtures, solutions of 0.1% w/v of porcine gelatin with carrageenan were prepared in the presence of sodium chloride or calcium chloride with a salt concentration of 0 to 200 mM in the final solution, after which pH-turbidimetric titration with acetic acid and sodium hydroxide was carried out for each solution.

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CONFLICT OF INTEREST

The authors declare no conflict of interest.

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