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STRUCTURE AND PHYSICO-MECHANICAL PROPERTIES OF POLYELECTROLYTE COMPLEXES BASED ON SODIUM CARBOXYMETHYLCELLULOSE POLYSACCHARIDE AND POLYACRYLAMIDE

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In this paper, the structure and physico-mechanical properties of films of polyelectrolyte complexes (PEC) based on sodium carboxymethylcellulose (Na-CMC) with linear polyacrylamide (PAA) have been studied. Polyelectrolyte complexes were obtained by mixing aqueous solutions of Na-CMC and PAA components in various ratios of components and pH of the medium. The structure of the obtained products was determined using IR spectroscopy and electron microscopy. IR spectra in the range 400–4000 cm^{-1} were recorded on NIKOLET Magna-560 IR and Specord-75IR spectrophotometers (Carl Zeiss, GDR). The mechanical properties of films of polyelectrolyte complexes were determined by stretching at a constant speed of movement of the lower clamp, 50 mm/min, on an Instron-1100 automatic dynamometer (England) at room temperature. IR spectroscopic data showed that polyelectrolyte complexes based on Na-CMC and PAA were stabilized due to the cooperative ionic bond between Na-CMC carboxylate anions ($-COO^-$) and amine groups ($-NH_2$) of polyacrylamide. It is shown that PEC films with an equimolar ratio of Na-CMC and PAA components have an increased value of mechanical strength (σ_p = 38 MPa), elastic modulus (E = 73 MPa) and a minimum relative elongation (ε = 0.5%). And in excess of Na-CMC or PAA leads to a decrease in mechanical strength and elastic modulus, which is associated with a decrease in the frequency of intermolecular bonds. It has been ascertained that water-soluble polyelectrolyte complexes based on Na-CMC and PAA with increased strength properties can be obtained from solutions of components taken at an equimolar ratio of interacting components. By changing the ratio of components, properties such as mechanical strength, modulus of elasticity and elongation can be controlled. This can serve as one of the means of controlling the structure and properties of Na-CMC and PAA polyelectrolyte complexes. The regulation of the physico-mechanical properties of PEC films opens up wide opportunities for their use as a soil structure former in agriculture and water management and as the basis for soft drugs in pharmacy.
Keywords: *Sodium carboxymethylcellulose; Polyacrylamide; Polycomplex; Interpolymer complex; Films; Structure; Properties; Mechanical strength; Relative elongation; Modulus of elasticity; Electron microscopy*
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INTRODUCTION

Currently, of great scientific and practical interest is the study of the ability of many water-soluble natural and synthetic polymers to form stable products of cooperative reactions between heterogeneous polymers, called polyelectrolyte complexes (PEC) [1–3]. PEC is a new individual substance formed due to the interaction of a polycation and a polyanion with the formation of an ionic bond. In terms of solving this problem, the interaction products of natural polyelectrolytes that form PECs are of the greatest interest. PECs are promising products in pharmacy and are increasingly used as thickeners and stabilizers for suspensions, prolongers of the drugs action, and film formers for capsules and tablets, as a base for ointments and other soft dosage forms, since they exhibit a number of unique and most valuable properties [4-11].

The possibility of independent variation of formation conditions, the ratio of reagents, and their molecular structure is essentially the basis for controlling the process of formation and production of IPC with a given set of properties [12-16].

Very interesting and promising in this aspect are macromolecular complexes based on sodium carboxymethylcellulose (Na-CMC) (polyanion) and linear polyacrylamide (PAA) (polycation), which form polyelectrolyte complexes (PEC) in aqueous systems. The structure of a natural cellulose polysaccharide derivative, Na-CMC, the presence of polar-functional groups in its macromolecules, which cause intense intermolecular interactions, the high degree of orientation of this rigid-chain polymer, determines its ability to exhibit the properties of a matrix carrier and a complexing agent with linear polyacrylamide.

Of great interest is the unique feature of the PEC systems structure based on Na-CMC and PAA systems, where, as a result of self-organization of macromolecules during the formation of polycomplexes, nanostructures with controlled nanosizes are formed [17]. This feature of polycomplex systems creates the possibility of molecular penetration of their drug compounds at the cellular level, which allows them to be used as drug systems carriers with directed transport properties. The consistency and normality of the structural and mechanical parameters of the bases based on IPC provides optimal bioavailability of medicinal substances, easy, painless application of the ointment to the skin, mucous membranes, etc. [18]. It should be noted that the constituent components of polycomplexes (PC) and PEC are large-tonnage, accessible, and cheap polymers of local origin [19].

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In scientific terms, the interest in these objects is explained by the complexity of the complex structure of systems of these polymers, and in practical terms, these studies are relevant due to the possibility of using them in pharmacy as bases for ointments and soft dosage forms [20, 21].

In this regard, this paper is devoted to the study of the structure and physico-mechanical properties of PEC obtained on the basis of Na-CMC with PAA of a linear structure with varying the ratio of the interacting components and depending on the pH of the medium.

Of undoubted interest is the study of the physico-mechanical properties of IPC films, since these properties are directly related to the structure of the polymer body and the possibility of their application [2, 22, 23]. Such a study is of independent scientific value and practical importance, since the structure of the Na-CMC-PAA PEC can be varied by changing the ratio of the interacting components, and the mechanical properties directly depend on both the structure of the initial components and the structure of the PEC, and largely determine the areas of possible their use in the national economy. In the literature, studies in this direction are few, and the interest in the study of the physico-mechanical properties of Na-CMC-PAA PEC films is due to the possibility of obtaining new polymeric materials with desired mechanical properties and their targeted use as bases for soft drugs in pharmacy [24] and as a soil structure former in agriculture and water management [21,25].

EXPERIMENTAL PART

Materials

Sodium carboxymethylcellulose (Na-CMC) - SSt 5.588-79 and BA 6-05-386-80. Purified sodium carboxymethyl cellulose, a product of the Namangan Chemical Plant, obtained by the method of heterogeneous solid-phase esterification of sulfite wood cellulose with monochloroacetic acid (MCA) of the following structure was used as the main object of the research:

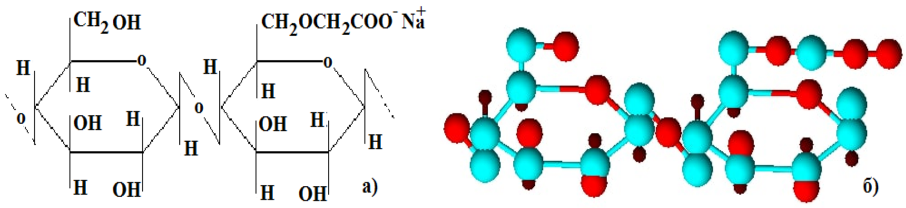

Figure 1. Chemical formula (a) and model structure of Na-CMC (b)

with a degree of substitution (DS) - 70 and a degree of polymerization (DP) - 450. It was repeatedly purified from low molecular weight salts according to the procedure given in [26, 27].

Na-CMC is a weak polyacid, its dissociation constant depends on the C3. When the DS changes from 10 to 80, the dissociation constant changes from $5.25\cdot10^{-7}$ to $5\cdot10^{-5}$. In practice, CMC is mainly used in the form of the Na salt.

Polyacrylamide (PAA). The second component of the polyelectrolyte complex is polyacrylamide, a product of the Navoi Chemical Plant according to TSh 6.1-00203849-64:1997. Polyacrylamide is a polyelectrolyte obtained on the basis of the polymerization of acrylamide containing the element nitrogen, which has a linearly branched structure (Fig. 2) [28-31].

Polyacrylamide is an odorless, amorphous solid with a white-yellowish color, the molecular weight of which is 10^4-10^7 (depending on the conditions of preparation). The density of polyacrylamide at room temperature (295-297 K) is about 1.302 g/cm^3. The decomposition temperature is about 463 K. Polyacrylamide is a polyelectrolyte with hygroscopic properties, harmless, and forms a soft gel when dissolved in water.

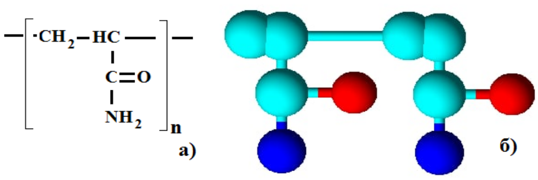

Figure 2. Chemical formula (a) and model structure of PAA (b)

Synthesis of PEC based on Na-CMC with polyacrylamide

We used solutions of Na-CMC in bidistilled water with a concentration of 0.01 to 0.4 basic mol/l. Reaction mixtures of the required concentrations were prepared by mixing reagent solutions in the appropriate proportion at room temperature and pH 6.0–7.2.

Obtaining films of polyelectrolyte complexes

Films from polyelectrolyte complexes were obtained by mixing aqueous solutions of Na-CMC and PAA components in equinormal ratios at different contents of components and pH of the medium. The solutions were poured onto an optical glass substrate and evaporated at room temperature. Solid dry films were washed with bidistilled water to neutral pH, then dried at room temperature.

IR spectroscopic studies

The structure of the obtained products was determined using IR spectroscopy and electron microscopy. IR spectra in the range 400–4000 cm^{-1} were recorded on "NIKOLET Magna-560 IR" and "Specord-75IR" spectrophotometers (Carl Zeiss, GDR). Samples for IR spectroscopy were prepared in the form of pellets with KBr, films on a KRS-5 plate, and films 8–12 μm thick. Films on a KRS-5 plate were obtained by evaporation of the solvent (water) at room temperature (295–297 K).

Study of mechanical properties

The mechanical properties of the polyelectrolyte complexes films were determined by stretching at a constant lower clamp speed of 50 mm/min on an Instron-1100 automatic dynamometer (England) and on a Shimadzu AGS-X universal tensile testing machine at room temperature. The maximum measurement error was 1% [25]. Samples of the studied films were prepared in the form of blades with a working area of 5x50 mm and a thickness of 0.07 mm. Samples were measured in an air-dry state, pre-conditioned at a certain air humidity. Tensile stress σ_p (MPa) in uniaxial tension was calculated by the formula:

$$\sigma_p = \frac{P}{S_0} \quad (1)$$

where, *P*- is the breaking force acting on the sample; S_0- is the initial cross section of the sample.

Relative elongation was calculated by the formula:

$$\varepsilon = \frac{l - l_0}{l_0}.100\% \quad (2)$$

where, l_0 and l are the lengths of the original and stretched samples, respectively.

The initial modulus of elasticity was calculated on the initial straight sections of the stretching curves using the formula:

$$E_0 = \frac{\sigma}{\varepsilon} \quad (3)$$

Viscometric properties

The viscosity of solutions of polyelectrolyte complexes was determined on an Ubbellod viscometer (d = 2 mm), at various temperatures under thermostatic conditions, and the time of solution outflow from the capillary was determined. The technique for determining the viscosity of solutions is described in detail in the paper [32].

Electron microscopic studies

Electron microscopic studies of the surfaces and cleavages (ends) of PEC films were carried out on a "Hitachi-520" scanning electron microscope (Japan) with a resolution of 60 Å. Samples were obtained by the brittle cleavage method at liquid nitrogen temperature [33]. The research results were recorded on electron micrographs.

RESULTS AND DISCUSSION

When mixed under certain technological conditions and at a certain temperature, and also, in principle, from different polyelectrolytes with different structures, a new, individual substance was obtained, which differed both in properties and in structure from the original components shown in Fig. 3.

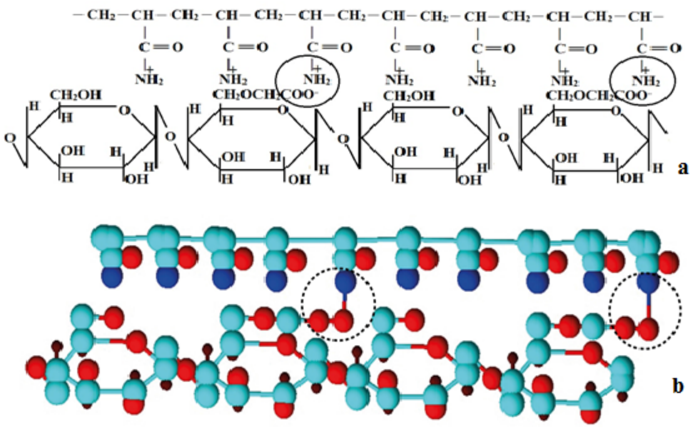

Figure 3. Chemical formula (a) and model structure (b) of the polyelectrolyte complex obtained on the basis of Na-CMC and PAA. Shaded area shows the formation of ionic bond between Na-CMC and PAA

Experimental data on the study of the solutions interaction of Na-CMC and polyacrylamide in neutral and slightly acidic media showed that when mixing solutions, a change in the pH of the polyelectrolyte complex compared to the initial components (Fig. 4). It should be noted that the initial components of the polyelectrolyte complex have a pH in the region of neutral values. The initial addition of polyacrylamide to the Na-CMC solution results in a slight change in the pH reading to an equimolar composition. A further increase in the proportion of the polyacrylamide solution leads to a sharp increase in the pH of the solution of the polyelectrolyte complex. A sharp break in the dependence of the pH of solutions on the ratio of components corresponds to the equimolar composition of the interacting components (Fig. 4).

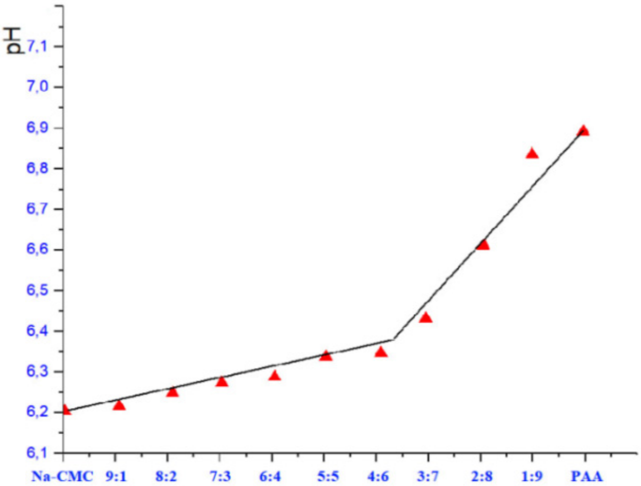

Figure 4. The dependence of the pH value of polyelectrolyte complexes on the ratio of Na-CMC and PAA components

The above data are confirmed by data on the study of electrical conductivity and viscosity of solutions of polyelectrolyte complexes from the ratio of Na-CMC:PAA components (table 1). A comparative change in the pH readings, electrical conductivity and viscosity of solutions of polyelectrolyte complexes from additivity indicates the formation of new individual substances compared to the original components.

Table 1. Physico-chemical and electrical properties of PEC based on Na-CMC and PAA

PEC Properties	Na-CMC : PAA ratio										
	Na-КМЦ	9:1	8:2	7:3	6:4	5:5	4:6	3:7	2:8	1:9	ПАА
Viscosity, η, Pa·s	0.71	0.71	0.72	1.03	1.56	2.01	2.32	2.38	2.39	2.20	2.13
Electrical conductivity, σ, Cm	186	272	314	390	451	518	575	633	715	770	793

The structure of the resulting PEC based on Na-CMC and polyacrylamide was ascertained by IR spectroscopy and on the basis of literature data [29, 30]. The results of the analysis of the IR spectrum of the initial product Na-KMC showed that the IR spectrum contains such functional groups (Fig. 5) as the carboxylate anion - COO^- (1603 cm^{-1} and 1419 cm^{-1}), the carboxyl group - COOH and the hydroxyl group is OH, as well as the presence of absorption bands in the structure at 1650, 1550, 1400, 1250, 1020, 780 cm^{-1}, which gives these polymers polyelectrolyte properties (Table 2, Fig. 5). The IR spectrum of the second component contains absorption bands characteristic of a linearly branched structure. Analysis of the IR spectrum of polyacrylamide showed that the structure has absorption bands at 3422, 3180, 1664, 1618, 1401, 1327, 1112, 618 cm^{-1}, which exhibits the characteristic properties of a polyelectrolyte (Table 2, Fig. 6).

Table 2. Absorption band of functional groups in Na-CMC and PAA macromolecules and in PEC

Na-CMC	Na-CMC: PAA, 80:20	Na-CMC: PAA, 60:40	Na-CMC: PAA, 50:50	Na-CMC: PAA, 40:60	PAA
3443.2	3442.52	3441.83	3429.20	3429.20	3422.96
2923.09	2923.49	-	3205.15	3207.46	3180.47
-	2854.49	-	-	-	-
-	1664.79	1668.27	1667.71	1667.42	1664.71
1603.76	1609.92	1609.46	1609.89	1612.41	1618.95
1419.79	1414.20	1452.46	1451.46	1452.41	1401.76
-	-	1411.60	1407.45	1409.54	
1270.80	1326.20	1325.32	1324.72	1324.85	1327.14
1114.64	1266.51	1265.71	1117.50	1112.88	1112.51
1060.75	1151.51	1150.79	1066.41	-	-
1022.15	1125.71	1123.39	-	-	-
-	1060.57	1060.30	-	-	-
-	1022.79	-	-	-	-
915.74	915.79	914.13	914.72	915.69	
701.87	621.73	622.02	619.58	617.32	618.63
599.17	-	-	-	-	-

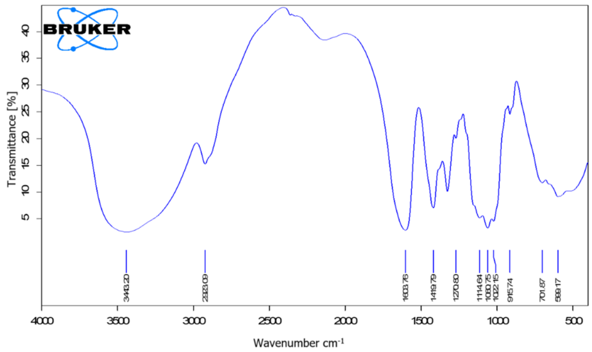

Figure 5. IR spectra of Na-CMC

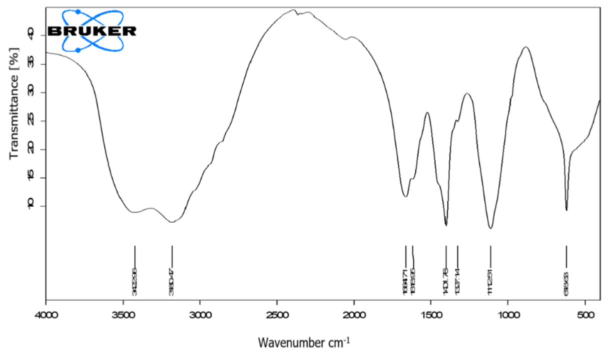

Figure 6. IR spectra of polyacrylamide

The change in intensity and the shift of the absorption band in the area of 1667 cm^{-1}, 1609 cm^{-1} and in the area of 1407 cm^{-1}, 1451 cm^{-1} indicates the formation of an ionic bond between the carboxylate anions of Na-CMC and the amine groups of polyacrylamides, which gives this polyelectrolyte complex new properties that differ from the original components (Fig. 7).

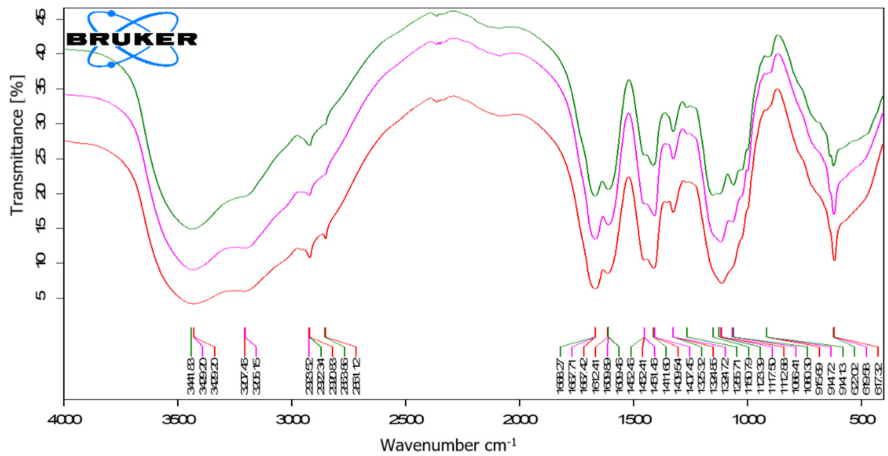

Figure 7. IR spectra of polyelectrolyte complexes from the ratio of components Na-CMC and PAA: 1 - 60:40; 2- 50:50; 3-40:60

Thus, the results of IR spectroscopy of the polyelectrolyte complex obtained on the basis of Na-CMC and polyacrylamide in neutral and slightly acidic media showed that water-soluble polyelectrolyte complexes are formed upon mixing. The resulting product can be used as carriers for soft drugs.

The study of the physico-mechanical properties of polyelectrolyte complex films obtained on the basis of Na-CMC and PAA undoubtedly confirmed the formation of a new individual substance. Since the physico-mechanical properties of such films are related to the structure of the obtained new individual polyelectrolyte complexes, their use in engineering and technology and for the preparation of drugs with prolonged properties, as well as in other substrates, is of great importance. Such studies are of both scientific and practical importance; the physico-mechanical properties and structure of the polyelectrolyte complex films will depend on the structure and properties of its constituent components [34, 35].

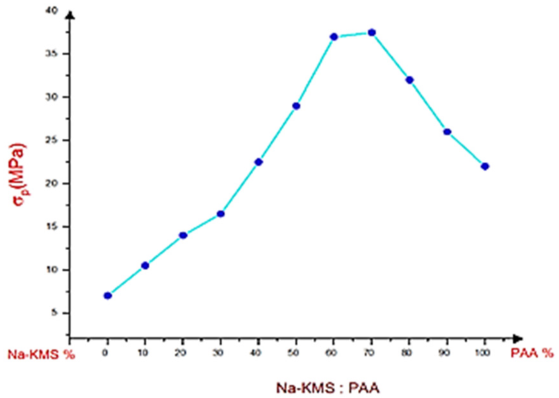

Figure 8. Graph of the dependence of the films strength of polyelectrolyte complexes on the ratio of Na-CMC:PAA components. Temperature T=24°C

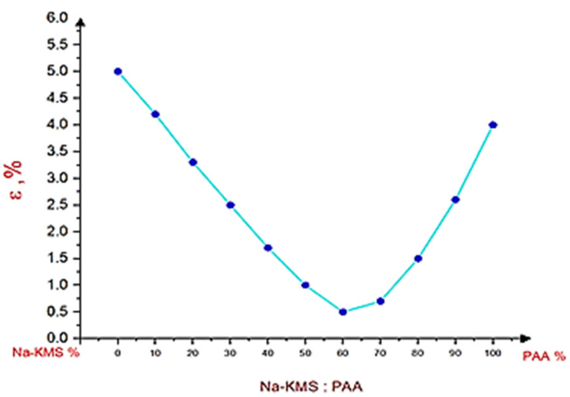

Figure 9. Graph of the dependence of the films elasticity of polyelectrolyte complexes on the ratio of Na-CMC:PAA components. Temperature T=24°C

The physico-mechanical characteristics of the polyelectrolyte complexes films were studied on an Instron-1100 automatic dynamometer (England) and on a Shimadzu AGS-X universal tensile testing machine at room temperature by stretching with a constant lower clamping speed of 50 mm/min. The mechanical strength, relative elongation and modulus of elasticity of the polyelectrolyte complexes films based on Na-CMC and PAA were determined (Fig. 8-10).

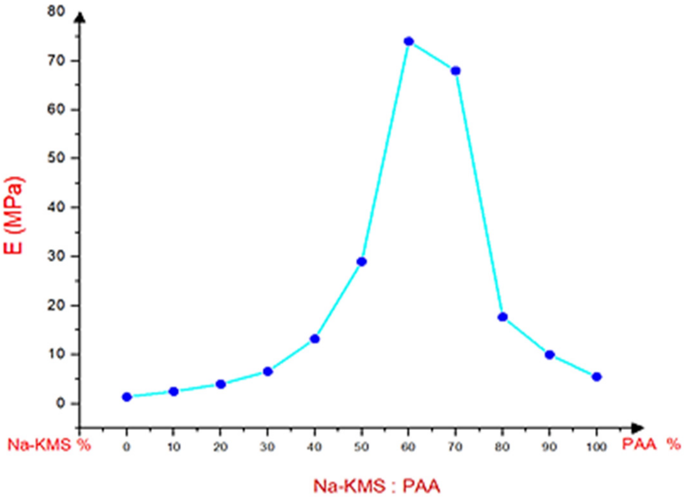

Figure 10. Graph of the dependence of the films elastic modulus of polyelectrolyte complexes on the ratio of components Na-CMC : PAA. Temperature T=24°C

The results of the experimental data showed that with an increase in the content of PAA in the composition of polyelectrolyte complexes obtained on the basis of Na-CMC and PAA, the interaction between the constituent components of the polyelectrolyte complex increases, which leads to an increase in the mechanical strength of the films to an equimolar composition (Fig. 8). In this case, one can observe a decrease in the relative elongation of films of polyelectrolyte complexes (Fig. 9). By increasing the frequency of crosslinking between the constituent components of the polyelectrolyte complex, the elasticity modulus of the films increases, which can be traced from the graph of the dependence of the elasticity modulus of the polyelectrolyte complexes films on the ratio of Na-CMC: PAA components to the equimolar composition (Fig. 10). A further increase in PAA in the composition of polyelectrolyte complexes leads to a decrease in the mechanical strength of the films (Fig. 8), an increase in relative elongation (Fig. 9) and a decrease in the elastic modulus (Fig. 10).

A further increase in the content of PAA in the composition of the polyelectrolyte complex films and, therefore, one can observe a decrease in the mechanical strength of the films due to an increase in the dispersed phase (Fig. 8), an increase in the relative elongation of the films under tension (Fig. 9) and a decrease in the elastic modulus due to an increase in the heterogeneous structure in films of the polyelectrolyte complex (Fig. 10). This is mainly due to the formation of a heterogeneous structure and, as a result, a decrease in the frequency of intermolecular bonds. In addition, a decrease in the elasticity modulus in films of polyelectrolyte complexes with an excess of Na-KMC or PAA is associated with the formation of a heterogeneous structure, which can be observed from images obtained by scanning electron microscopy (Fig. 11).

Thus, the resulting polyelectrolyte complex based on Na-CMC and PAA is a new, individual substance, both in structure and in properties differing from the structure and properties of the original products. It should be noted that the pH value for Na-CMC is pH = 6.2; for PAA - pH = 7.0, and for PEC with an equimolar composition, it has pH = 6.4. Mechanical strength for Na-CMC - σ_p = 6.1 MPa; for PAA, σ_p = 22 MPa, and for PEC films with an equimolar composition, it has σ_p = 38 MPa. Relative elongation for Na-CMC - ε = 5.1%; for PAA – ε = 4.0%, and for PEC films with equimolar composition it has ε=0.5%. The modulus of elasticity of films for Na-CMC is E = 3.2 MPa; for PAA – E = 8.1 MPa, and for PEC films with an equimolar composition is E = 73 MPa. All these changes in the properties of films of polyelectrolyte complexes are associated with the formation of a cooperative ionic bond between the carboxylate anions of Na-CMC (COO^-) and the amine groups of polyacrylamide, which was confirmed by IR spectroscopic data. The increase in mechanical strength (σ_p) and elastic modulus (E) and the decrease in the relative elongation of the films of polyelectrolyte complexes is associated with an increase in the density of crosslinking between the constituent components of the polyelectrolyte complex. It should be noted that the obtained films at an equimolar ratio of Na-CMC : PAA components have the maximum mechanical strength (σ_p), elastic modulus (E), and minimum relative elongation (ε).

CONCLUSIONS

1. Based on the experimental data, it can be concluded that a new individual substance based on Na-CMC and PAA has been obtained. The structure and properties of the obtained polyelectrolyte complex differ from the initial products.

2. From the data of IR spectroscopy, it was revealed that the polyelectrolyte complex based on Na-CMC and PAA is stabilized due to the ionic bond between the carboxylate anions of Na-CMC and the amine groups of polyacrylamides.

3. Ionic bonds largely determine the mechanical strength of the systems under study. Water-soluble polyelectrolyte complexes based on Na-CMC and PAA with increased strength properties can be obtained from solutions of components taken at an equimolar ratio of interacting components. By changing the ratio of components, properties such as mechanical strength, modulus of elasticity and elongation can be controlled. This can serve as one of the means of controlling the structure and properties of Na-CMC and PAA polyelectrolyte complexes.

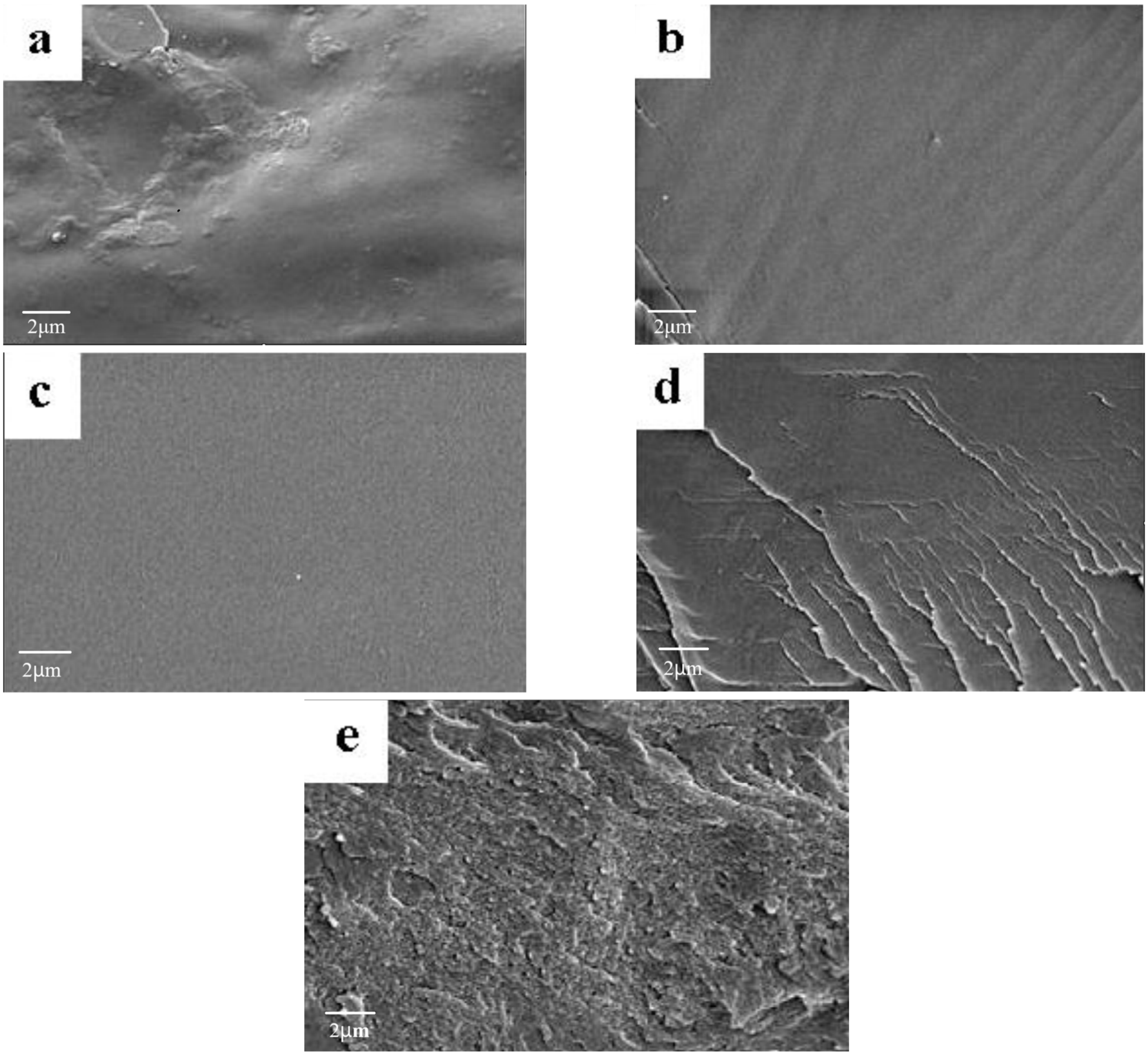

Figure 11. SEM electron microscopy images of Na-CMC (a), PAA (b) and PEC with the ratio of components Na-CMC : PAA=1:1 (c); 2:1(d); 1:2(e).

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СТРУКТУРА ТА ФІЗИКО-МЕХАНІЧНІ ВЛАСТИВОСТІ ПОЛІЕЛЕКТРОЛІТНИХ КОМПЛЕКСІВ НА ОСНОВІ ПОЛІСАХАРИДУ КАРБОКСИМЕТИЛЦЕЛЮЛОЗИ НАТРІЮ ТА ПОЛІАКРИЛАМІДУ

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У роботі досліджено структуру та фізико-механічні властивості плівок поліелектролітних комплексів (ПЕК) на основі натрійкарбоксиметилцелюлози (Na-КМЦ) з лінійним поліакриламідом (ПАК). Поліелектролітні комплекси отримували змішуванням водних розчинів компонентів Na-КМЦ і ПАА в різних співвідношеннях компонентів і pH середовища. Структуру отриманих продуктів визначали за допомогою ІЧ-спектроскопії та електронної мікроскопії. ІЧ-спектри в діапазоні 400–4000 см$^{-1}$ записували на спектрофотометрах NIKOLET Magna-560 IR та Specord-75IR (Carl Zeiss, НДР). Механічні властивості плівок поліелектролітних комплексів визначали розтягуванням при постійній швидкості руху нижнього затискача 50 мм/хв на автоматичному динамометрі Instron-1100 (Англія) при кімнатній температурі. Дані ІЧ-спектроскопії показали, що поліелектролітні комплекси на основі Na-КМЦ і ПАА стабілізувалися за рахунок кооперативного іонного зв'язку між карбоксилат-аніонами Na-КМЦ (-COO-) і амінними групами (-NH2) поліакриламіду. Показано, що плівки ПЕК з еквімолярним співвідношенням компонентів Na-КМЦ та ПАА мають підвищене значення механічної міцності (σ_p = 38 МПа), модуля пружності (E=73 МПа) та мінімальне відносне видовження (ε = 0,5%). А надлишок Na-CMC або PAA призводить до зниження механічної міцності і модуля пружності, що пов'язано зі зниженням частоти міжмолекулярних зв'язків. Встановлено, що з розчинів компонентів, узятих при еквімолярному співвідношенні взаємодіючих компонентів, можна отримати водорозчинні поліелектролітні комплекси на основі Na-КМЦ і ПАА з підвищеними міцнісними властивостями. Змінюючи співвідношення компонентів, можна контролювати такі властивості, як механічна міцність, модуль пружності та подовження. Це може бути одним із засобів керування структурою та властивостями поліелектролітних комплексів Na-КМЦ та ПАА. Регулювання фізико-механічних властивостей плівок ПЕК відкриває широкі можливості для їх використання як ґрунтоутворювача у сільському та водному господарстві та як основи для м'яких лікарських засобів у фармації.

Ключові слова: *натрійкарбоксиметилцелюлоза; поліакриламід; полікомплекс; інтерполімерний комплекс; плівки; структура; властивості; механічна міцність; відносне видовження; модуль пружності; електронна мікроскопія*