

FEATURES OF THE INTERACTION OF INTERGEL SYSTEMS BASED ON POLYACRYLIC ACID AND POLY-2-METHYL-5-VINYLPYRIDINE WITH GOLD IONS

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Abstract. *Introduction.* In the interpolymer system of polyacrylic acid and poly-2-methyl-5-vinylpyridine, the change in the mutual activation of polymer hydrogels with different degrees of crosslinking as a result of remote interaction of the two hydrogels was determined, and their effect on the process of gold ion sorption was studied [1,2]. The purpose aim of the work is to study the sorption capacity of intergel systems based on poly-2-methyl-5-vinylpyridine and polyacrylic acid in relation to gold ions [3,4]. *The obtained results* The maximum degree of sorption of the gPAA:gP2M5VP interpolymer systems increased to 0.645 mol/L at a polymer molar ratio of 5:1 within 48 hours of interaction, and then gradually decreased to 0.575 mol/L. The maximum pH values of the intergel system gPAA:gP2M5VP are observed at a ratio of 3:3, corresponding to a pH of 2.99. The minimum pH value is 2.96 at a ratio of 2:4. The overall swelling ratio (Ksw) increases from 0.8 to 3 during the conformational rotation of the interstitial unit. *Conclusion.* According to the results, the electrochemical properties of hydrogels of polyacrylic acid and poly-2-methyl-5-vinylpyridine in the interpolymer system were studied and the optimal molar ratios were determined. Thus, as a result of these studies: the obtained experimental data on specific electrical conductivity of the solution, pH of the medium and swelling coefficient of hydrogels showed that the presence of remote interaction between acrylic acid and 2-methyl-5-vinylpyridine hydrogels has a significant effect on the initial polymer network and physicochemical properties of the solution.

Keywords: interpolymer system, ion-exchangers, hydrogels, polyacrylic acid, poly-2-methyl-5-vinylpyridine, remote interaction, mutual activation, sorption.

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1. Introduction

In recent years, work that has attracted the attention of scientists and has been intensively studied is primarily focused on the separation of gold ions from solutions by creating interpolymer systems of cross-linked polymers with high sorption properties and selectivity, as well as on the determination and analysis of their sorption capacity [5,6]. A distinctive feature of the ionization process in interpolymer systems is the absence of a counterion in one part of the ionized chains. This is a consequence of interpolymer interactions, leading to mutual activation of polymers and the formation of unbalanced charges along the polymer chain. During the dissociation of acidic polymers, a proton is separated from the carboxyl group, and this ion combines with a heteroatom of the base polymer in an aqueous medium, resulting in the formation of a neutralized charge. In this case, the charge density of the base polymer is limited by the degree of dissociation of the acidic polymer. As a result, the polyacid undergoes ionization, followed by dissociation of the carboxyl group, and then the association of protons with the polybase heteroatom, resulting in both polymers having equally charged groups in the interstitial chain links without counterions. Therefore, interpolymer systems have a higher sorption capacity compared to individual polymers [7-9]. Based on the long-distance interaction, hydrogels of polyacrylic acid (PAA) and poly-2-methyl-5-vinylpyridine (P2M5VP) were selected for the formation of an intergel pair in intergel systems. Purpose of the study: poly-2-methyl-5-vinylpyridine and polyacrylic acid. Study of the sorption capacity of intergel systems based on gold ions.

2. Materials and Methods

The study was conducted at room temperature over various time intervals. The interpolymer system was prepared as follows: each dry mass of hydrogel was weighed and placed separately into specially prepared individual polypropylene cells (filters) permeable to molecular ions and molecules, but not to hydrogel dispersions. Electrochemical changes (specific conductivity, pH) of the aqueous solution were measured at various time intervals. The mass of the empty cell (g) was subtracted from the mass of the cell filled with hydrogel. gPAA:gP2M5VP. The swelling degree of the hydrogels was calculated. We poured 200 ml of the prepared solution into seven glasses and added seven different ratios of gPAA:gP2M5VP. We conducted a study by placing a cell containing gels in an intergel system. A polyacrylic hydrogel was synthesized in the presence of the cross-linking agent N,N-methylene-bis-acrylamide and the $K_2S_2O_8$ - $Na_2S_2O_3$ redox system. Poly(2-methyl-5-vinylpyridine) hydrogel (gP2M5VP) was synthesized from the linear polymer in a dimethylformamide medium in the presence of epichlorohydrin at 60°C. The degree of purification was monitored using a conductivity meter. During the study, the synthesized poly(2-methyl-5-vinylpyridine) hydrogel was washed with distilled water for two weeks, dried for one week, and used for experimental work. Conductometer MARK 603 for determining the electrical conductivity of aqueous solutions. The study was

conducted by Expert 002 (Econix-Expert, Moscow, Russian Federation). To measure pH values, use pH Lab 827 (Metrohm, Heritau, Objects (Switzerland) were used. The study of the electrochemical properties of polymers allows us to predict the formation of highly ionized regions of polymer structures during long-range interaction.

3. Results and discussion

Based on our systematic work, we first synthesized the required acidic hydrogels based on polyacrylic acid (gPAA) and poly-2-methyl-5-vinylpyridine (gP2M5VP) and analyzed their properties using various methods. Having created an intergel system, we studied the selective adsorption of the gold ion HAuCl_4 in various gPAA:gP2M5VP ratios. Previous studies have shown that hydrogels interact with each other at a distance in various combinations, changing their electrochemical and volumetric-hydrodynamic properties [10]. To create an intergel system by mixing poly (2-methyl-5-vinylpyridine) gel with poly(acrylic acid) in different ratios, we first studied the changes in electrical conductivity, pH, and swelling coefficient of the synthesized poly (acrylic acid). Figure 1 shows the changes in the specific electrical conductivity of the medium over certain time intervals at different molar ratios of the gPAA:gP2M5VP intergel system. We observe that at ratios of 6:0 and 5:1 in the intergel system, the specific electrical conductivity of the medium reaches its maximum value, suggesting mutual activation of the hydrogels and their transition to a highly ionized state based on long-distance interactions. The maximum degree of sorption of the gPAA:gP2M5VP intergel system is observed at a molar ratio of polymers of 5:1 over 48 hours of interaction. We see that the concentration of gold ions increases to 0.645 mol/L and then gradually decreases to 0.575 mol/L. The figure also shows high conductivity values at a hydrogel ratio of 4:2, which is 0.625 mol/L. The maximum values observed at the top point indicate an increase in the concentration of electrical conductivity charge carriers, which is explained by the maximum formation of ionic bonds and the corresponding increase in electrical conductivity as a result of the optimal number of carboxyl and amino groups. High values of specific electrical conductivity of the intergel system gPAA:gP2M5VP in a ratio of 1:5. Indicates the presence of H^+ ions in an aqueous medium. The concentration of H^+ ions depends on the degree of dissociation of the carboxyl groups. The presence of a maximum on the curve indicates that the speed of ion movement in dilute solutions of strong electrolytes does not depend strongly on concentration. In a solution of $\text{HAuCl}_4 \cdot 4\text{H}_2\text{O}$. Changes in pH of the environment are shown in Figure 2.

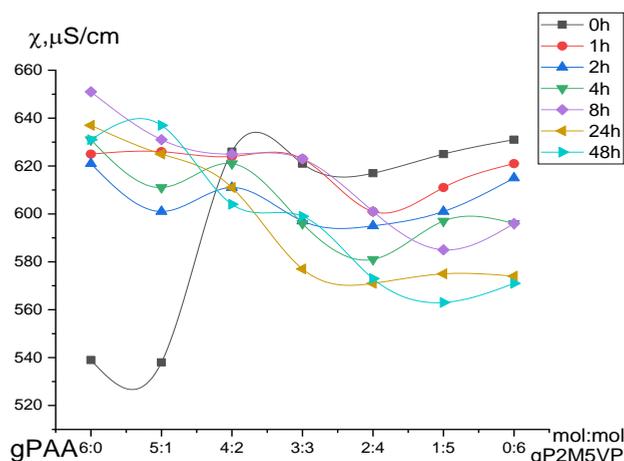


Figure 1 – Dependence of parameters of intrinsic electroconductivity of gPAA:gP2M5VP hydrogels with different ratios on molar ratios.

The results of the study show that the minimum and maximum pH values are observed in the $\text{HAuCl}_4 \cdot 4\text{H}_2\text{O}$ solution. The maximum pH point of the medium is close to neutral at a ratio of 6:0, $\text{pH}_{\text{max}} = 3.03$. The minimum pH values of the gPAA:gP2M5VP intergel system medium are observed at ratios of 4:2 and 1:5, and the maximum values are at ratios of 5:1 and 3:3, respectively, at a ratio of 3:3, $\text{pH}_{\text{max}} = 2.99$. These values indicate that a low pH value indicates an increase in the concentration of hydrogen ions, and a high pH value indicates a low concentration of H^+ ions. At a ratio of 4:2, $\text{pH}_{\text{max}} = 2.98$, and at a ratio of 2:4, $\text{pH}_{\text{max}} = 2.96$. These values indicate that the concentrations of H^+ and OH^- ions in the solution are close. In a $\text{HAuCl}_4 \cdot 4\text{H}_2\text{O}$ solution, the pH of the medium slowly increased at a ratio of 2:4, and then the hydrogen concentration remained constant, indicating the formation of hydroxyl ions as a result of the interaction of the base with water molecules in the solution. In the intergel system, at an equimolar ratio of 3:3, complete neutralization of H^+ and OH^- ions occurs. At this ratio, the electrical conductivity of the system is minimal. At a ratio of 1:5, the pH decreases, and the electrical conductivity reaches its maximum. As can be seen from the figure, the low pH and high electrical conductivity are due to the influence of H^+ ions. gPAA:gP2M5VP The dependence of the swelling coefficients (K_{sw}) of the intergel system on different time intervals is shown in Figure 3. The swelling ratio of the gPAA hydrogel remains constant at a ratio of 6:0 over different time intervals. This indicates that the hydrogel has a limited capacity to absorb water, as only one polymer, gPAA, is present in this system.

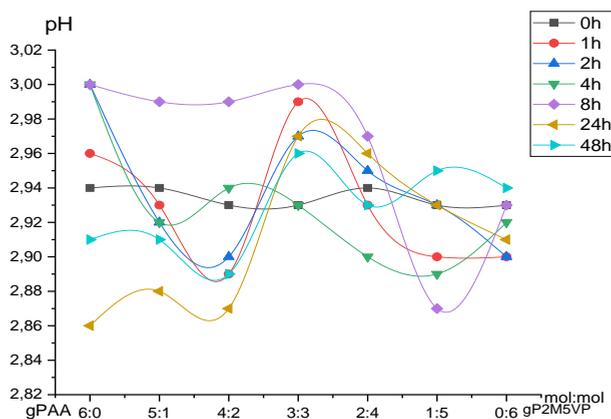


Figure 2 - Dependence of pH of the intergel system of polyacrylic acid and poly-2-methyl-5-vinylpyridine on the molar ratios of hydrogels.

The swelling coefficients of the hydrogels were $K_{sw}(\text{gPAA})=10.1$ and $(\text{gP2M5VP}) = 0.46$. These data indicate a high degree of ionization of both hydrogels at this stage. The concentration of H^+ and OH^- ions is proportional to the concentration of the hydrogels and their degree of swelling. In the $\text{HAuCl}_4 \cdot 4\text{H}_2\text{O}$ solution, at a certain ratio of acidic to basic hydrogels, we observed the highest swelling coefficient (K_{sw}) at a ratio of 1:5 after 48 hours compared to the initial hydrogels, indicating a high degree of ionization of the hydrogel at this stage.

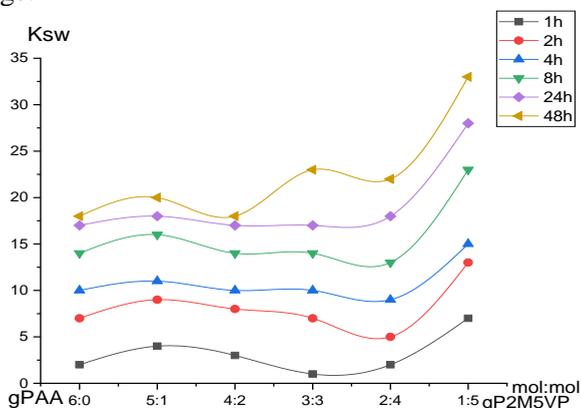


Figure 3– Dependence of the degree of swelling of gPAA in the presence of hydrogel P2M5VP on the molar ratio of hydrogels.

The high degree of H^+ ionization is explained by the excess of polyacid in the solution and the high dissociation of COOH groups. The figure shows that the swelling coefficient (K_{sw}) increases with time, and after a certain time, swelling

ceases. The swelling coefficient (K_{sw}) of the polyacrylic acid hydrogel in the intergel system in the presence of poly(2-methyl-5-vinylpyridine) demonstrates that the volumetric and gravimetric properties of the hydrogels change during long-distance interaction. A sharp increase in the size of the poly(2-methyl-5-vinylpyridine) gel is observed in the ratios of 4:2 and 3:3. As a result of long-distance interaction in the $\text{HAuCl}_4 \cdot 4\text{H}_2\text{O}$ solution, a sharp decrease in the swelling coefficient is observed in the ratio of 3:3, which is due to an insufficient swelling rate and low concentrations of the main groups.

gPAA:gP2M5VP Figure 4 shows the dependence of the swelling degree (K_{sw}) of poly-2-methyl-5-vinylpyridine gel on time at different time intervals in the intergel system. Unlike the cross-linked hydrogel gPAA, the hydrogel P2M5VP exhibits a stepwise change in the swelling degree (K_{sw}) over time. The maximum swelling of the gel gP2M5VP is observed at ratios of 5:1 and 4:2, which is evident from the change in the swelling coefficient of poly-2-methyl-5-vinylpyridine gel with a certain time interval, which increases to a minimum value of 0.1 and a maximum value of 2.1, as evidenced by the change in the volumetric-gravimetric properties of poly-2-methyl-5-vinylpyridine hydrogel and polyacrylic acid hydrogel.

The maximum swelling values of the hydrogels are also maintained at a ratio of 2:4. This is explained by the neutralization of hydroxyl ions by protons and a very low degree of dissociation of carboxyl groups while maintaining a high concentration of positive ions in the solution. gPAA:gP2M5VP hydrogels in the intergel system showed a minimum value at a ratio of 4:2 in a $\text{HAuCl}_4 \cdot 4\text{H}_2\text{O}$ solution, which is due to the insufficient swelling rate of the main groups and their low concentration.

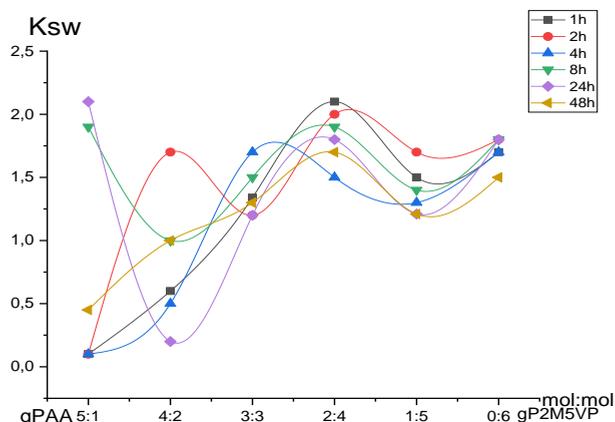


Figure 4 – Dependence of the degree of swelling of gP2M5VP in the presence of gPAA hydrogel on the molar ratio of hydrogels.

Thus, the poly(2-methyl-5-vinylpyridine) hydrogel changes its volumetric and gravimetric properties compared to the polyacrylic acid hydrogel. Overall,

(K_{sw}) increases from 0.8 to 3 during the conformational rotation of the internodal unit (K_{sw}). That is, the swelling coefficient increases by 3.8 times. The minimal reduction in swelling in this region is associated with the formation of an intramolecular bond of the $\geq NH^+ \dots N \leq$ type or the formation of an intramolecular association. As the gPAA concentration in the aqueous medium increases, the swelling process intensifies, and K_H increases significantly. In this case, the local conformational change and the inverse arrangement of interstitials are explained by an increase in chain charge density, which leads to the rupture of intramolecular bonds.

4. Conclusion

The results of numerous studies and analyzes, depending on the amount and volume of cross-linked polymers used in the synthesis, show that mutual activation of interpolymer systems of polyacrylic acid and poly-2-methyl-5-vinylpyridine leads to a significant increase in their sorption capacity. Experimental data on the specific electrical conductivity of the solution, pH of the medium and the swelling coefficient of the hydrogels showed that the presence of long-range interactions between acrylic acid and 2-methyl-5-vinylpyridine hydrogels has a significant effect on the initial polymer network and the physicochemical properties of the solution; the presence of areas of abnormally high electrical conductivity at the lowest gPAA:gP2M5VP ratio indicates the presence of highly concentrated charged ions; in an equimolar gPAA:gP2M5VP ratio, association of H^+ and OH^- ions with each other and intermolecular duplication of hydrogels occurs; As a result of long-distance interaction, additional activation of the hydrogels occurs, i.e., the interstitial chain acquires an additional charge without a negative ion; long-distance interaction leads to a conformational change in the interstitial chain in both hydrogels, which causes their additional swelling; at a certain ratio of gPAA:gP2M5VP, a zone of intramolecular association formation was identified in the gP2M5VP hydrogel, which leads to rotation of the polymer network.

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ПОЛИАКРИЛ ҚЫШҚЫЛЫ ЖӘНЕ ПОЛИ-2-МЕТИЛ-5-ВИНИЛПИРИДИН ГИДРОГЕЛЬДЕРІ НЕГІЗІНДЕ ҚҰРЫЛҒАН ИНТЕРГЕЛЬДІ ЖҮЙЕЛЕРДІҢ АЛТЫН ИОНДАРЫНА ҚАТЫСТЫ ӘРЕКЕТТЕСУ ЕРЕКШЕЛІКТЕРІ

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Түйіндеме. *Кіріспе.* Полиакрил қышқылы мен поли-2-метил-5-винилпиридин интерполимерлік жүйесінде екі гидрогелдің қашықтықтан әрекеттесуі нәтижесінде торлану дәрежесі әр түрлі полимерлік гидрогелдердің өзара активтенуінің өзгерісі анықталып, олардың алтын иондарының сорбциялау үдерісіне тигізетін әсері зерттелді [1,2]. *Жұмыстың мақсаты* поли-2-метил-5-винилпиридин және полиакрил қышқылы негізінде құрылған интергельді жүйелердің алтын иондарына қатысты сорбциялық қабілетін зерттеу [3,4]. *Алынған нәтижелер.* гПАК:гП2М5ВП интерполимерлі жүйелерінің максималды сорбция дәрежесі 48 сағат әрекеттесу кезінде полимерлердің 5:1 молярлық қатынастарында алтын иондарының концентрациясы 0.645 моль/л мөлшеріне дейін жоғарлап, ары қарай біртіндеп 0.575 моль/л азайған. ПАК:П2М5ВП интергельді жүйесінің рН ортасының максималды мәндері 3:3 қатынасында байқалады, сәйкесінше рН 2.99 тең. Ал минимум 2:4 қатынасында рН мәні 2.96 тең. Жалпы ісіну дәрежесінің (K_i) түйінаралық буын конформациялық айналу үрдісінде (K_i) 0.8 ден 3-ке дейін өседі. *Қорытынды.* Көптеген зерттеулер мен талдаулардың нәтижелері синтездеу барысындағы пайдаланған сызықтық полимерлермен ондағы қосымша реагенттердің мөлшеріне, көлеміне қарай алынған мәліметтер полиакрил қышқылы мен поли-2-метил-5-винилпиридин интерполимерлі жүйелерінің өзара активтенуі олардың сорбциялық қабілетінің айтарлықтай жоғарылауына әкелетінін көрсетеді.

Түйін сөздер: интерполимерлік жүйе, иониттер, полиакрил қышқылы, поли-2-метил-5-винилпиридин, қашықтықтан өзара әрекеттесу, гидрогельдер, өзара активтену, сорбция.

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ОСОБЕННОСТИ ВЗАИМОДЕЙСТВИЯ ИНТЕРГЕЛЕВЫХ СИСТЕМ НА ОСНОВЕ ПОЛИАКРИЛОВОЙ КИСЛОТЫ И ПОЛИ-2-МЕТИЛ-5-ВИНИЛПИРИДИНА С ИОНАМИ ЗОЛОТА

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Резюме. *Введение.* В результате дистанционного взаимодействия двух гидрогелей в интерполимерной системе полиакриловой кислоты и поли-2-метил-5-винилпиридина было определено изменение взаимной активации полимерных гидрогелей с различной степенью сшивания, а также изучено их влияние на процесс сорбции ионов золота [1,2]. *Целью работы* было изучение сорбционной способности интергелевых систем на основе поли-2-метил-5-винилпиридина и полиакриловой кислоты по отношению к ионам золота [3,4]. *Полученные результаты.* Максимальная степень сорбции интерполимерных систем гПАК:гП2М5ВП увеличилась до 0.645 моль/л при молярном соотношении полимеров 5:1 в течение 48 часов взаимодействия, а затем постепенно снизилась до 0.575 моль/л. Максимальные значения рН среды интергелевой системы гПАК:гП2М5ВП наблюдались при соотношении 3:3, соответственно, рН 2,99. А при минимальном соотношении 2:4 значение рН составляет 2,96. Общая степень набухания (K_n) увеличивается с 0.8 до 3 в процессе конформационного вращения междоузловое звена. *Вывод.* По результатам исследований изучены электрохимические свойства гидрогелей полиакриловой кислоты и поли-2-метил-5-винилпиридина в интерполимерной системе и определены оптимальные молярные соотношения. Таким образом, в результате проведенных исследований: полученные экспериментальные данные по удельной электропроводности раствора,

pH среды и коэффициенту набухания гидрогелей показали, что наличие дистанционного взаимодействия между акриловой кислотой и 2-метил-5-винилпиридином гидрогелей оказывает существенное влияние на исходную полимерную сетку и физико-химические свойства раствора.

Ключевые слова: интерполимерная система, иониты, гидрогели, полиакриловой кислота, поли-2-метил-5-винилпиридин, дистанционное взаимодействие, взаимная активация, сорбция.

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