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Specific features of uranyl ions extraction by interpolymer system based on polyacrylic acid and polyethyleneimine hydrogels

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ABSTRACT

Uranyl ions sorption of by interpolymer system consisting of polyacrylic acid hydrogel (hPAA) and polyethyleneimine hydrogel (hPEI) has been studied. Rate of uranyl ions extraction by the initial polymers and interpolymer system hPAA-hPEI, polymeric chain binding rate and dynamic exchange capacity of initial polymers and interpolymer system hPAA-hPEI were calculated. Based on obtained outcomes it was found that area of maximum rate of uranyl ions extraction is within the ratios of 67%hPAA:33%hPEI and 33%hPAA:67%hPEI. Maximum uranyl ions extraction rate after 48 hours of hydrogels remote interaction was 90.0 %, when polymeric chain binding rate was 9.1 % and dynamic exchange capacity was 1.14 mmol/g. Rate of uranyl ions extraction by the initial polymer hydrogels 100 % hPAA and 100 % hPEI was 68.0 % and 52.0%. Obtained outcomes showed changes of initial polymeric hydrogels sorption properties in intergel system leading to functional groups obtaining higher reactive ability, which makes it possible to use them for further development of highly efficient uranyl ions extraction sorption technology.

Keywords: hydrogels, interpolymer systems, sorption, polyacrylic acid, polyethylenimine, uranyl ion.

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Introduction

Rare-earth elements are used in multiple areas of contemporary engineering: radio electronics, instrumentation, nuclear science, machinery, chemical industry, metallurgy etc., which is why production and consumption of these elements is continuously growing worldwide. Rare-earth element containing ores extraction is often complicated by their radioactivity which is caused by them containing uranium, thorium and their half-life products. There is development of traditional raw material processing technologies as well as search for new ones in order to make mining of rare-earth elements containing ores

cheaper and produce high-purity individual elements [1-3].

Due to this fact, the issue of rare-earth elements separation from radioactive elements, particularly uranium, is of high importance nowadays [4-6].

Uranium can be removed from solutions by sorption, extraction and other methods. Sorption methods are more preferred nowadays than extraction ones. Sorption methods are more eco-friendly and have fewer technological cycles than extraction technologies [7-9].

Unfortunately, currently developed ion exchange resins cannot be widely applied; also, not all salt and metal ions have complementary

polymeric sorbents. Due to composition diversity of solutions extracted from different deposits it is difficult to study sorption process in model solutions and develop effective sorbents. Use of interpolymer systems allows highly efficient mass extraction of various elements' anions and cations [10-12].

Due to this, the objective of our research was to study depth of uranyl ions sorption during their interaction with interpolymer systems and prospects of using these interpolymer systems for uranium extraction from product solutions.

Experimental part

Equipment. Sorbent mass was determined by weighing at electronic analytical scales MSE125P-100-DO Sartorius Cubis (Germany).

Materials. The research was performed in water environment and in solution of hexaqua uranyl nitrate (UO_2^{2+} concentration = 100 mg/l) as well as polyacrylic acid and polyethylenimine hydrogels. Polyacrylic acid hydrogels were synthesized in presence of linking agent N, N - methylen-bis-acrylamide and red-ox system $\text{K}_2\text{S}_2\text{O}_8 - \text{Na}_2\text{S}_2\text{O}_3$ in water environment. Polyethylenimine hydrogels were synthesized with N, N - dimethylformamide solvent in presence of linking agent epichlorhydrin. Synthesized PAA and PEI hydrogels were crushed into small fractions of $250 < d < 425 \mu\text{m}$. Hydrogel swelling rates were $\alpha_{(\text{hPAA})} = 10,36 \text{ g/g}$, $\alpha_{(\text{hPEI})} = 8,36 \text{ g/g}$.

Intergel couple of polyacrylic acid hydrogel and polyethylenimine hydrogel (hPAA-hPEI) was combined for research purposes.

Experiment. Experiments were done at room temperature. PAA and PEI were taken in dry state. Study of the intergel systems was done as follows: each hydrogel in dry state was placed in separate polypropylene cages. Then polypropylene cages with dry hydrogels were placed in glasses with water and hexaqua uranyl nitrate solutions.

Research of individual polymeric hydrogels sorption properties was done as follows:

1) Estimated amount of each hydrogel in dry state was placed into polypropylene cages.

2) Uranyl ions sorption by individual PAA and PEI hydrogels was done for 48 hours. During this period aliquots were taken for further determination of uranyl ions concentration.

The procedure of studying intergel system sorption abilities is the same as that of studying individual polymer hydrogels.

Uranyl ions determination procedure.

Measurement of uranium mass concentration was done by volumetric titanium-phosphate-vanadate method based on red-ox properties of uranium (IV) and uranium (VI) and its reduction and oxidation reactions.

Uranyl ions extraction (sorption) rate was calculated according to the formula:

$$\eta = \frac{C_{in} - C_{res}}{C_{in}} * 100\%, \quad (1)$$

where C_{in} – uranyl ions initial concentration in solution, mg/l; C_{res} – uranyl ions residual concentration in solution, mg/l.

Total polymer chain binding rate was calculated according to the formula:

$$\theta = \frac{v_{sor}}{v} * 100\%, \quad (2)$$

where v_{sor} – amount of sorbed uranyl ions, mole (calculated as $v_{sor} = m/M$, where m – mass of sorbed uranyl ion, g, M - molecular mass of uranyl ion, g/mole); v – polymer weighed portion (calculated as $v = m/M$, where m – mass of PAA and PEI polymers, g, M - molecular mass of mass of PAA and PEI polymers, g/mole, if two hydrogels are present in the solution, this is calculated as summed amount of each of them), mole.

Dynamic exchange capacity of individual hydrogels and intergel system was calculated according to the formula:

$$Q = \frac{v_{sor}}{m_{sorbent}}, \quad (3)$$

where v_{sor} – amount of sorbed uranyl ions, mole (calculated as $v_{sor} = m/M$, where m – mass of sorbed uranyl ion, g, M - molecular mass of uranyl ion, g/mole); m – polymer weighed portion (if two hydrogels are present in the solution, this is calculated as summed weight of each of them), g.

Results discussion

Previously performed studies [10-12] have shown that almost all interpolymer systems based on acidic (polyacrylic and polymethacrylic acids) and basic (poly-4-vinylpyridine and poly-2-methyl-5-pyridine) lightly cross-linked polymeric hydrogels display higher activity than their initial components. It was also found that polymer ratios which provide higher ions sorption differ significantly depending

on nature of acidic and basic hydrogels and nature of rare-earth metals.

These results were obtained after studying sorption of lanthanum, cerium, dysprosium, neodymium, samarium and erbium by intergel systems. Influence of initial hydrogel state on sorption process was also found. Depending on what gel is used for intergel couple formation (dry, swollen or partially swollen) there can be different hydrogel ratios in intergel systems with high sorption capacity and rare-earth metal ions sorption rate.

However, no work was done to define optimal conditions for maximum sorption and ions selectivity in case of using intergel systems for uranyl ions extraction.

Main feature of intergel systems is absence of direct contact between polymer hydrogels in solution. In other words, this can be called a distant interaction of hydrogels. The general outline of intergel system is given at Figure 1.

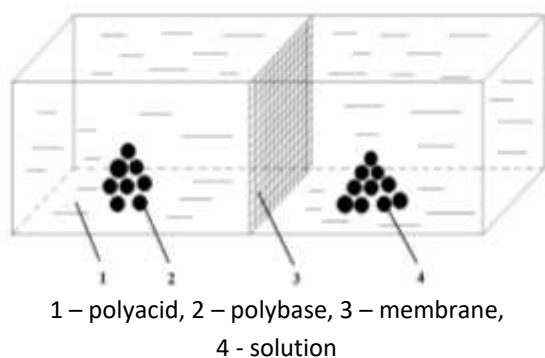
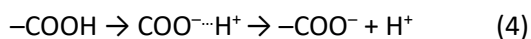


Figure 1 – Intergel system outline

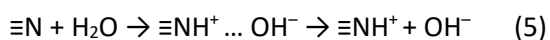
The following chemical reactions occur during distant hydrogel interaction:

1) Dissociation of $-COOH$ – groups of internode links:



It should be considered that first there is ionization with formation of ion couples, which later partially dissociate into separate ions.

2) Atom of nitrogen in pyridine ring is ionized and partially dissociated:



3) Then atom of nitrogen also interacts with proton separated from carboxylic group:



4) H^+ and OH^- ions appearing as a result of functional groups interaction with water molecules form water molecules (true for equimolar concentration of protons and hydroxyl ions):



According to equation 4, carboxylic groups are dissociated into carboxylate anions and protons depends on dissociation rate. As a result of proton adjoining by heteroatoms of nitrogen, total amount of hydrogen atoms in solution decreases, which leads to further dissociation of other (previously non-dissociated) functional carboxylic groups. This happens due to Le Chatelier principle because of equilibrium shift to the right (towards proton formation).

These interactions lead to uncompensated like-charged functional groups formation at internode links of both hydrogels (acidic and basic); these groups bounce off each other according to laws of electrostatics and lead to macromolecular coil expansion. The end result of these electrostatic interactions is significant swelling of polymer macromolecules.

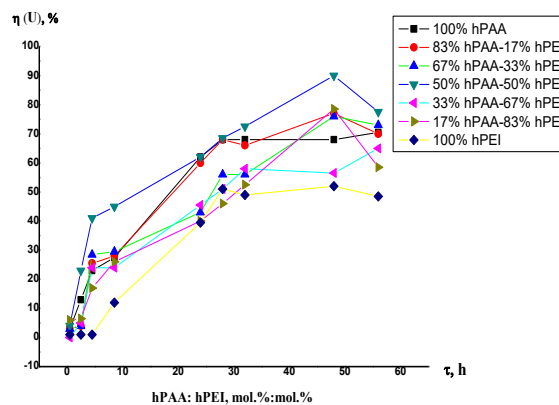


Figure 2 – Dependence of uranyl ions extraction rate by hPAA-hPEI intergel system over time

Figure 2 shows dependence of uranyl ions extraction rate by initial hydrogels and hPAA-hPEI intergel system over time. As the figure suggests, intergel system has increased uranyl ions extraction rate over time. It should be noted that polymeric macromolecules switching to highly ionized state due to hydrogels mutual activation during their remote interaction leads to significantly increased uranyl ions extraction rate with polymeric hydrogels in intergel couples in comparison with initial hydrogels. Most of uranyl ions are sorbed by initial PAA hydrogel and intergel systems through

48-hour interaction with salt solutions. Highest uranyl ions extraction rate is observed in intergel system within 67%hPAA:33%hPEI and 33%hPAA:67%hPEI ratios 48 hours later and is 90,0%. Uranyl ions extraction rate by initial polymer hydrogels - 100%hPAA and 100%hPEI is 68,0% and 52,0%, respectively. Initial PEI hydrogel provides only slight increase of sorption rate over time, however, hPEI (even with low sorption activity towards uranyl ions) actively participates in initial PAA hydrogel activation, which can be seen better within 67%hPAA:33%hPEI and 33%hPAA:67%hPEI ratios.

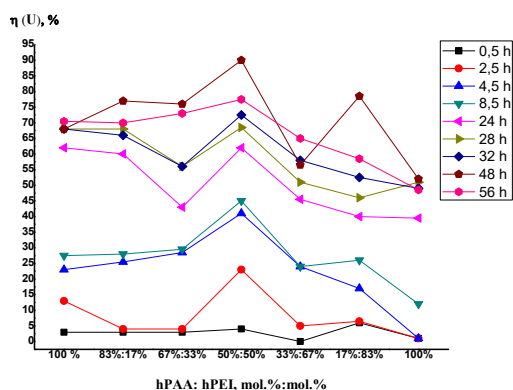


Figure 3 – Dependence of uranyl ions extraction rate by hPAA-hPEI intergel system on hydrogel molar ratio over time

Figure 3 shows dependence of uranyl ions extraction rate by initial hydrogels and hPAA-hPEI intergel system on hydrogel molar ratio over time.

As the figure suggests, the highest uranyl ions sorption occurs within 67%hPAA:33%hPEI and 33%hPAA:67%hPEI ratios. The highest uranyl ions extraction rate after 48 hours of distant hydrogel interaction is 90,0%. Primary reason for such high extraction rate is high polymeric structures ionization due to their mutual activation.

Table 1 shows polymeric chain binding rates (in relation to uranyl ions) by initial polymers and hPAA-hPEI intergel systems over time. The most intensive uranyl ions binding by initial polymers and intergel systems occurs within 48 hours. High degree of polymeric chain binding in relation to uranyl ions is observed at 67%hPAA:33%hPEI and 33%hPAA:67%hPEI components ratios and is 9,1 %. This suggests high macromolecules ionization rate due to mutual activation of PAA and PEI polymers. Individual PAA and PEI polymeric chain binding rate in relation to uranyl ions over 48 hours is 6,8 % and 5,3 %, respectively.

Table 2 shows dynamic exchange capacity (in relation to uranyl ions) by initial hydrogels and hPAA-hPEI intergel systems over time. Obtained data suggest that polymeric hydrogels mutual activation in intergel couples leads to significant increase of exchange capacity values in comparison with initial PEI hydrogel. This is the most distinctive after 48 hours of remote interaction. Intergel system reaches maximum values of dynamic exchange capacity within the ratios of 67%hPAA:33%hPEI and 33%hPAA:67%hPEI after 48 hours of remote interaction and is 1.14 mmol/g.

Table 1 – Polymeric chain binding rates (in relation to uranyl ions) by initial hydrogels and hPAA-hPEI intergel systems over time, %

Polymer chain binding rate (in relation to uranyl ions), %								
τ , h	0,5	2,5	4,5	8,5	24	28	32	48
100%hPAA	0,3	1,3	2,3	2,8	6,3	6,8	6,8	6,8
83% hPAA:17% hPEI	0,3	0,4	2,6	2,8	6,1	6,8	6,7	7,8
67% hPAA:33% hPEI	0,3	0,4	2,8	2,9	4,3	5,7	5,7	7,7
50% hPAA:50% hPEI	0,4	2,3	4,2	4,6	6,3	6,9	7,3	9,1
33% hPAA:67% hPEI	0,0	0,5	2,4	2,4	4,6	5,2	5,8	5,7
17% hPAA:83% hPEI	0,6	0,7	1,7	2,6	4,1	4,7	5,3	8,0
100%hPEI	0,0	0,1	0,1	2,1	4,0	5,2	4,9	5,3

Table 2 – Dynamic exchange capacity (in relation to uranyl ions) by initial hydrogels and hPAA-hPEI intergel systems over time, %

τ , h	0,5	2,5	4,5	8,5	24	28	32	48
100%hPAA	0,04	0,18	0,32	0,38	0,87	0,95	0,95	0,95
83% hPAA:17% hPEI	0,04	0,05	0,35	0,38	0,81	0,92	0,89	1,04
67% hPAA:33% hPEI	0,04	0,05	0,37	0,38	0,56	0,73	0,73	0,99
50% hPAA:50% hPEI	0,05	0,29	0,52	0,57	0,78	0,86	0,92	1,14
33% hPAA:67% hPEI	0,00	0,06	0,29	0,29	0,56	0,62	0,71	0,69
17% hPAA:83% hPEI	0,07	0,08	0,20	0,31	0,47	0,54	0,62	0,93
100%hPEI	0,01	0,01	0,01	0,12	0,45	0,58	0,56	0,60

Conclusion

Based on obtained results it was found that the highest uranyl ions sorption by interpolymer system occurs at 67%hPAA:33%hPEI and 33%hPAA:67%hPEI components ratio. The highest uranyl ions extraction rate after 48 hours of distant hydrogel interaction is 90,0%, where polymeric chain binding rate is 9,1% and dynamic exchange capacity is 1,14 mmole/g. Uranyl ions extraction rate by individual polymer hydrogels - 100%hPAA and 100%hPEI - is 68,0% and 52,0%, respectively. Due to mutual hydrogel activation during their distant interaction polymeric macromolecules

switch to highly ionized state which leads to significant increase of uranyl ions extraction rate in comparison with initial polymers. Obtained outcomes suggest that interpolymer systems can be used for highly efficient sorption technology of extracting uranyl ions and other elements from commercial solutions as well as for concentration, separation of different ions from water solutions for performing technological, environmental and other objectives.

Conflict of interest. On behalf of all authors the correspondent author declares that there is no conflict of interest.

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Уранил иондарын полиакрил қышқылы мен полиэтиленимин гидрогельдерінің негізіндегі интерполимерлік жүйемен бөлу ерекшеліктері

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ТҮЙІНДЕМЕ

Полиакрил қышқылы (ПАҚ) және полиэтиленимин (ПЭИ) гидрогелдерінен тұратын интерполимерлік жүйелердің уранил иондарына сорбциясы зерттелді. Бастапқы полимерлер мен ПАҚ-ПЭИг интерполимерлік жүйелерінің уранил иондарын бөлу дәрежесі, полимер тізбегінің байланысу дәрежесі және тиімді динамикалық алмасу сымдылығы есептелді. Алынған нәтижелер негізінде интерполимерлік жүйелердің уранил иондарына максималды сорбциясы

<p>Мақала келді: 14 шілде 2021 Сараптамадан өтті: 07 тамыз 2021 Қабылданды: 15 қазан 2021</p>	<p>67%ПАҚ:33%ПЭИ және 33%ПАҚ:67%ПЭИ қатынастарында болатыны анықталды. Гидрогелдердің қашықтан өзара әрекеттесуінің 48 сағатынан кейін уранил иондарын максималды бөлу дәрежесі 90,0 %, полимерлік тізбектің байланысу дәрежесі 9,1 % және динамикалық алмасу сиымдылығы – 1,14 ммоль/г болды. 100 % ПАҚ және 100 % ПЭИ жеке полимерлік гидрогелдерімен уранил иондарын бөлу дәрежесі сәйкесінше 68,0 % және 52,0 % құрады. Алынған нәтижелер интерполимерлі жүйелердегі бастапқы полимерлік гидрогелдердің сорбциялық қасиеттерінің өзгеретінін, яғни функционалды топтардың реакцияға түсу қабілеті анағұрлым артатынын көрсетті. Бұл оларды уранил иондарын бөлудің жоғары тиімді сорбциялық технологиясын жасау үшін қолдануға мүмкіндік береді.</p> <p>Түйін сөздер: гидрогелдер, интерполимерлі жүйелер, сорбция, полиакрил қышқылы, полиэтиленмин, уранил ионы.</p>
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Особенности извлечения ионов уранила интерполимерной системой на основе гидрогелей полиакриловой кислоты и полиэтиленмина

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<p>Поступила: 14 июля 2021 Рецензирование: 07 августа 2021 Принята в печать: 15 октября 2021</p>	<p>АННОТАЦИЯ Изучена сорбция ионов уранила интерполимерной системой, состоящей из гидрогеля полиакриловой кислоты (гПАК) и гидрогеля полиэтиленмина (гПЭИ). Были рассчитаны степень извлечения ионов уранила исходными полимерами и интерполимерной системой гПАК-гПЭИ, степень связывания полимерной цепи и динамическая обменная емкость исходных полимеров и интерполимерной системы гПАК-гПЭИ. На основе полученных результатов было установлено, что наибольшая степень сорбции ионов уранила интерполимерной системой происходит в пределах соотношений 67%гПАК:33%гПЭИ и 33%гПАК:67%гПЭИ. Максимальная степень извлечения ионов уранила по истечении 48 часов дистанционного взаимодействия гидрогелей составила 90,0 %, при которой степень связывания полимерной цепи составила 9,1 % и динамическая обменная емкость – 1,14 ммоль/г. Степень извлечения ионов уранила индивидуальными полимерными гидрогелями 100%гПАК и 100%гПЭИ составила 68,0 % и 52,0 % соответственно. Полученные результаты показали изменения сорбционных свойств исходных полимерных гидрогелей в интерполимерной системе, приводящие к тому, что функциональные группы преобретают более высокую реакционную способность, что предоставляет возможность их применения для последующей разработки высокоэффективной сорбционной технологии извлечения ионов уранила.</p> <p>Ключевые слова: гидрогели, интерполимерные системы, сорбция, полиакриловая кислота, полиэтиленмин, ион уранила.</p>
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