

NEWS

OF THE NATIONAL ACADEMY OF SCIENCES OF THE REPUBLIC OF KAZAKHSTAN

SERIES OF GEOLOGY AND TECHNICAL SCIENCES

ISSN 2224-5278

Volume 3, Number 423 (2017), 198 – 205

UDC 544.773.432:621.382-022.532

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REDISTRIBUTION OF THE CONCENTRATIONS IN POLYELECTROLYTE HYDROGELS CONTACTS AS THE BASIS OF NEW DESALINATION TECHNOLOGIES

Abstract. The possibility of using temperature-sensitive hydrogels for the demineralization of aqueous solutions of low molecular weight salts is presented in this article. In particular, the proposed method is applicable to saline groundwater, desalination problem, which is especially acute for many regions of Kazakhstan. The article shows that the use of temperature-sensitive hydrogels allows to use solar radiation energy for producing demineralized water. The proposed method is based on the concentrations redistribution effect, by which highly charged gel upon swelling in a saline solution absorbs water depleted by a low molecular weight component. In the next stage of the desalination cycle, the accumulated liquid is separated from the polyelectrolyte gel being in contact with the thermosensitive polymer network. At the final stage, desalinated liquid separates from the thermosensitive gel by heating at the sunlight. The intermediate stage is required because the direct heating of the polyelectrolyte gel does not provide its collapse, and the heat-sensitive gels alone do not have the density of the grid charge sufficient for concentrations redistribution effect. To implement this method of desalination, it is necessary to develop a technique of engineering calculation allowing to determine the characteristics of the heat-sensitive gel that is used for a particular range of concentrations. This, in turn, requires a generalization of previously developed theory of the concentrations redistribution effect in case of contact of the two grids with different degrees of ionization. Such theory is proposed in this article. It is based on the solution of equations describing the balance of low-molecular ions concentrations at the junction of two grids. It was found that the equations of this type can be solved analytically for the important in practice cases. These solutions meet the qualitative analysis of system behavior. Our theory results in the correspondence principle, i.e., the obtained solutions in the limiting case of the zero-degree ionization of one of the meshes transform to the results of the previously developed theory. This enables the use of the proposed theory as a basis for engineering calculation of temperature-sensitive hydrogels characteristics used in the desalination cycle.

Keywords: demineralized water, aqueous solutions, low-molecular ion, temperature-sensitive hydrogels, saline groundwater.

Desalination of natural water is a challenge for a number of Kazakhstan regions including the region where the capital is located. In the suburbs of Astana and surrounding areas, there are numerous sources of groundwater, but their use (e.g., for agricultural purposes) is hindered by the presence of low molecular weight salts, particularly by sodium chloride. At present, there are numerous methods of demineralization of aqueous solutions (reverse osmosis, electro dialysis, etc.), but such purified water in the above regions cannot be forwarded into economic turnover due to economic factors, primarily related to the energy expenses.

Authors of the article [1] proposed the method of desalting the liquids based on the use of concentrations redistribution effect [2, 3], which was observed when a cross-linked polyelectrolyte network was

placed into a low-molecular salt solution. If the concentration of salt in solution is 3-5 times lower than the concentration of ionized hydrogel groups (in terms of maximally swollen state), the polyelectrolyte network will absorb the depleted solution, the concentration of which is less than the initial one. In accordance to methodology [1], the depleted solution is separated from the hydrogel by the effect of electric current.

Thus, the method of demineralization of water (including groundwater) [1], consists of two stages: 1) the gel accumulates the depleted solution by the low-molecular weight component, and 2) this solution is separated from working substance by an electric current.

Expenses of energy necessary for separating the liquid from the hydrogel are quite high (they can be precisely estimated using an analogy with Faraday's law [4]). However, the second stage of this method can be realized by using thermo-sensitive hydrogels, which allows conducting demineralization directly using energy of solar radiation.

Indeed, the main property of thermo-sensitive hydrogels [5,6] is the ability to undergo through phase transition while increasing a temperature. This phase transition is characterized by rapid decrease in gel volume, accompanied by the separation of accumulated water. It is essential that this phase transition takes place under temperatures from 30 to 50°C. Consequently, it can be caused directly by solar radiation heating, which allows to exclude expenses on electric power for the second stage of desalination cycle.

However, the realization of this cycle being analogous to [1], in which the separation of demineralized water is carried out due to heating of the working substance by solar radiation is hampered by the specificity of the mechanism of phase transition that causes the collapse of thermo-sensitive hydrogels. Namely, this phase transition takes place in the network, which simultaneously includes both hydrophobic and hydrophilic functional groups [7]. The resulting degree of swelling is determined by the hydrophobic-hydrophilic balance, which can be shifted to one side or the other by external influences. In particular, an increase in temperature enhances hydrophobic interactions [7]; as a result, there is a phase transition of given type. Consequently, only hydrogels with a relatively low amount of hydrophilic functional groups can be thermo-sensitive. Otherwise, the hydrophobic-hydrophilic balance will be irreversibly shifted to the region of hydrophilic interactions. This means that thermo-sensitive polymer hydrogels can not have a significant density of the net charge because hydrophilic interactions are the most pronounced due to the dissociation of ionogenic groups.

This property does not allow using thermo-sensitive hydrogels directly in the desalination cycle, similar to the one described in the work [1]: that is why it is necessary to modify this approach. The technology of demineralization of water proposed in this article is based on the use of the following cycle. In the first stage [1], the polyelectrolyte hydrogel interacts with demineralized water. At the same time, due to the above mentioned effect of redistribution of concentrations, the swelling polymer network accumulates a solution concentration of which is much lower than the initial one.

In the second stage, the swollen polyelectrolyte hydrogel is brought into contact with a weakly charged thermo-sensitive network, which is in a non-swollen state. Due to the fact that in the volume of the polyelectrolyte hydrogel the concentration of the low molecular weight salt is relatively small (for the above mentioned reasons), the low density of its charge is no longer significant: some amount of the liquid accumulated by the polyelectrolyte gel passes into a weakly charged thermo-sensitive network. In the third stage, this liquid is separated from the thermo-sensitive gel by heating with solar heat.

Implementation of this methodology requires, among other things, the development in a methodology of calculating the characteristics of the used working substances. Therefore, there comes a problem of a theoretical description of the redistribution effect of the concentrations in a low-molecular salt that occurs upon contact of two cross-linked networks with different network-charge densities. This problem is solved in this article.

Let's write the Donnan balance equations in the following form, considering, for definiteness, the case where both networks are saturated with sodium chloride.

$$[Na^+]_i = \alpha [Na^+]_e \quad (1)$$

$$[Cl^-]_i = \alpha^{-1} [Cl^-]_e \quad (2)$$

where the chemical symbol in square parentheses represents the concentration of the corresponding ions, α - the Donnan coefficient, the indices i, e referring to the ion concentrations inside and outside the hydrogel, which has the maximum charge density, respectively.

Equations (1), (2), expressing Donnan's equilibrium, should be supplemented by ratios, presenting the neutrality condition in both networks.

$$[-COO^-]_e + [Cl^-]_e = [Na^+]_e \quad (3)$$

$$[-COO^-]_i + [Cl^-]_i = [Na^+]_i, \quad (4)$$

It is assumed that the density of both grids remains unchanged, and the degree of their dissociation is close to 100%. If there is no free solution in the system, the low-molecular component is distributed only between the two networks that are under consideration, i.e.

$$w[Cl^-]_i + (1-w)[Cl^-]_e = c_0 \quad (5)$$

where w is the fraction of the volume occupied by the gel with a higher net charge density in the system.

Ratio (5) expresses the constant condition of the total amount of low-molecular salt in the system under consideration. A similar condition for the concentration of cations should not be written down, since it is linearly dependent on the already presented relationships.

Equations (1) - (5) represent a closed system that allows to find the concentrations in both parts of the system if the net charge densities and the concentration of the initial solution are known.

We emphasize that the recorded system of equations is completely analogous to that used in [2, 8], the difference is that the presence of a charge in both contacting networks is taken into account. The results of the cited studies show that the calculation performed on the basis of the used equations satisfactorily describes the experimental data. This gives reason to suppose that the characteristics of the working substances used in the desalination cycle based on thermo-sensitive hydrogels can be obtained by calculation.

Substituting ratios (3) and (4) in (1), we immediately obtain a system of three algebraic equations:

$$[-COO^-]_i + [Cl^-]_i = \alpha([-COO^-]_e + [Cl^-]_e) \quad (6)$$

Substituting equations (3) and (4) into (1), we immediately obtain a system of three algebraic equations, which includes relation (6) and expressions (5) and (2). Equations (6) and (2) form a system of linear equations for the concentration of anions, which allows us to express these quantities through other ones. By substituting (2) into (6), we have

$$[-COO^-]_i + \alpha^{-1}[Cl^-]_e = \alpha([-COO^-]_e + [Cl^-]_e) \quad (7)$$

Where it comes to

$$[Cl^-]_e = \frac{[-COO^-]_i - \alpha[-COO^-]_e}{\alpha - \alpha^{-1}} \quad (8)$$

It also implies (2) that

$$[Cl^-]_i = \alpha^{-1}[Cl^-]_e = \frac{[-COO^-]_i - \alpha[-COO^-]_e}{\alpha^2 - 1} \quad (9)$$

Substituting (8) and (9) into relation (5), we obtain the only equation for the Donnan factor.

$$w \frac{[-COO^-]_i - \alpha[-COO^-]_e}{\alpha^2 - 1} + (1-w) \frac{[-COO^-]_i - \alpha[-COO^-]_e}{\alpha - \alpha^{-1}} = c_0 \quad (10)$$

Or

$$([\text{-COO}^-]_i - \alpha[\text{-COO}^-]_e) \left(\frac{w}{\alpha^2 - 1} + \frac{1-w}{\alpha - \alpha^{-1}} \right) = c_0 \quad (11)$$

Where it comes to

$$([\text{-COO}^-]_i - \alpha[\text{-COO}^-]_e) (w + (1-w)\alpha) = c_0 (\alpha^2 - 1) \quad (12)$$

It can be seen that this equation is quadratic relatively to Donnan's factor.

For more convenience, we put the marks:

$$N_i = [\text{-COO}^-]_i; \quad N_e = [\text{-COO}^-]_e \quad (13)$$

Then

$$(N_i - \alpha N_e) (w + (1-w)\alpha) = c_0 (\alpha^2 - 1) \quad (14)$$

or

$$\alpha^2 (c_0 + N_e (1-w)) + \alpha (N_e w - N_i (1-w)) - (c_0 + N_i w) = 0 \quad (15)$$

Equation (15) has, as expected, a well-defined symmetry. Namely, if we go from the factor α to the opposite of it

$$\tilde{\alpha} = \alpha^{-1} \quad (16)$$

i.e. to consider the symmetry case (respectively to the choice of the Donnan factor), then equation (15) goes into relation

$$(c_0 + N_e (1-w)) + \tilde{\alpha} (N_e w - N_i (1-w)) - \tilde{\alpha}^2 (c_0 + N_i w) = 0 \quad (17)$$

It is seen that if in the equation (17), the "i" and "e" indices are interchanged, and if relations are replaced

$$\tilde{w} = 1-w \quad (18)$$

Then it goes over into equation (15), which expresses the symmetry mentioned above.

The solution of the equation (15) has the form

$$\alpha = \frac{N_i (1-w) - N_e w + \sqrt{(N_e w - N_i (1-w))^2 + 4(c_0 + N_e (1-w))(c_0 + N_i w)}}{2(c_0 + N_e (1-w))} \quad (19)$$

In the expression (19), the plus sign is chosen at the root since only the positive values of the Donnan factor have a physical meaning.

For clarity, it can be rewritten using the reduced net charge density

$$n_i = \frac{N_i}{c_0}; \quad n_e = \frac{N_e}{c_0}, \quad (20)$$

We have

$$\alpha = \frac{n_i (1-w) - n_e w + \sqrt{(n_e w - n_i (1-w))^2 + 4(1+n_e (1-w))(1+n_i w)}}{2(1+n_e (1-w))} \quad (21)$$

Dependences of the potential on the parameter n_e less dense network for different values of w in two different values of n_i are presented in Figure 1a, b.

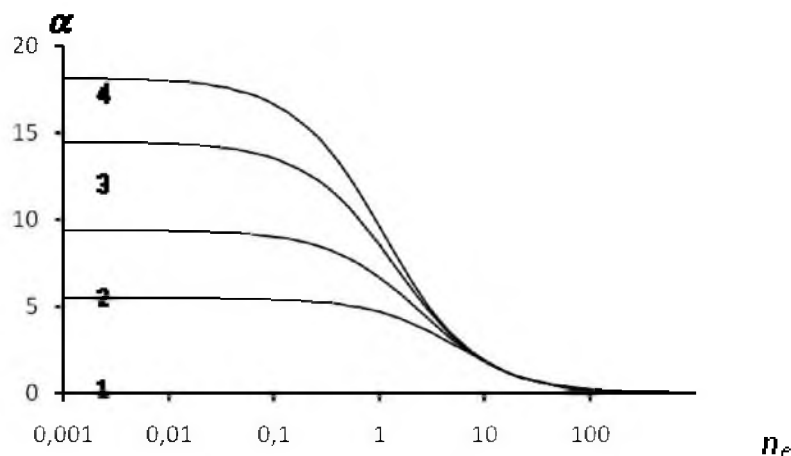


Figure 1a – Dependences of the Donnan factor α on the parameter n_e at $n_i = 20$ and different values of $w = 0.9$ (1); 0.6 (2); 0.3 (3); 0.1 (4)

It is seen that in the region $n_e < n_i$, $\alpha < 1$ holds, that is, as follows from (2), the concentration of anions (coinciding in the case under consideration with the concentration of a low molecular weight salt) in a more ionized network is lower than their concentration in a less ionized one. This completely corresponds to the conclusions of [2, 3].

Namely, the unequal value of the degree of ionization of the networks leads to a redistribution of the concentration of the low molecular weight salt, with the difference that, because of the nonzero degree of ionization in both networks, this effect becomes less pronounced compared with the case where the polyelectrolyte hydrogel is located in a solution of the low molecular weight salt.

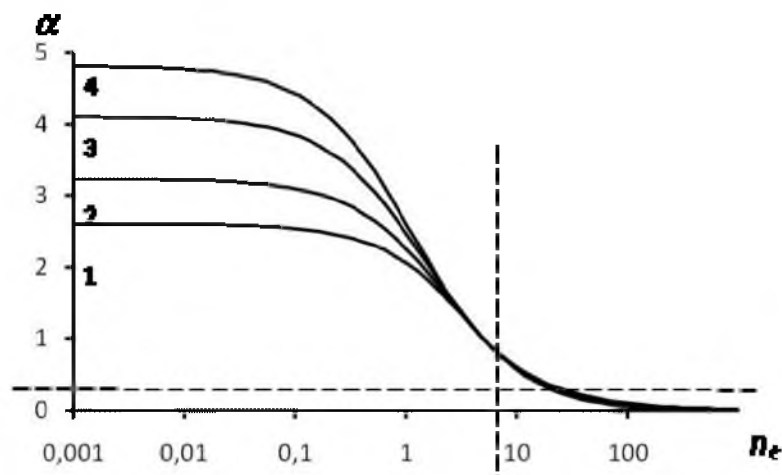


Figure 1b – Dependences of the Donnan factor α on the parameter n_e at $n_i = 20$ and different values: $w = 0.9$ (1); 0.6 (2); 0.3 (3); 0.1 (4)

This is also evident from Figure 1. Namely, with increasing n_e , the value of the factor α decreases, i.e. as the densities of the networks under consideration approach each other, the difference in the concentrations between the solutions accumulated by the two networks becomes less noticeable. More precisely, this conclusion is valid for the region $n_e < n_i$.

In the case when $n_e = n_i$, as expected, the value of α becomes merely equal to one. This is emphasized by Figure 1b, in which the corresponding point is marked with dashed lines.

With the inverse relation $n_e > n_i$, $\alpha < 1$ holds, which also indicates the existence of a concentration redistribution effect. However, in this case, the concentration of the low-molecular component in the network characterized by the parameter n_i , on the contrary, becomes larger than in the network characterized by the parameter n_e . In fact, the degree of ionization of the last network becomes larger.

Figure 1 also shows that in the region $n_e < n_i$, the value of α increases as the proportion of the volume falling on a denser grid decreases (as w decreases). This is quite understandable, since as w decreases, the volume increases, where a low-molecular component can be displaced.

Following [2, 3], we will use the given values of the concentrations

$$C_e = \frac{c_e}{N_i}; \quad n = \frac{N_e}{N_i} \quad (22)$$

Examples of the dependencies of the reduced concentration of a low molecular weight salt in a less dense network on the initial reduced concentration are shown in Figure 2. (These dependencies are constructed on the basis of the solution obtained for the Donnan factor.)

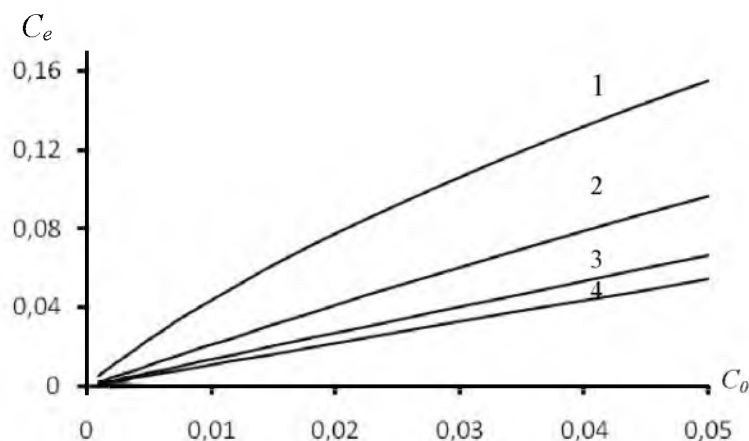


Figure 2 – Dependences of the reduced concentration of a low molecular weight salt in a less dense network C_e from the initial C_0 , range up to 0.05, $n = 0.1$, $w = 0.9$ (1); 0.6 (2); 0.3 (3); 0.1 (4); 0.05

It is seen that the concentration of a low-molecular component in a less dense network decreases as the fraction of the volume falling on a denser network decreases. This, in particular, shows that it is possible to ensure the operation of the second stage of the cycle in question, choosing the ratio of the volumes of polyelectrolyte and thermo-sensitive networks. Namely, by choosing a sufficiently large volume of a thermo-sensitive network (in comparison with the volume of a polyelectrolyte gel), one can choose conditions under which the concentration of the solution accumulated by this network will be several times lower than the initial one.

Thus, the advantage of the proposed technology is the direct use of solar energy for the demineralization of water. When this range of operating temperatures lies in the range of 30-500°C. This figure is easily achievable in solar heaters. There is also the possibility to provide a calculation of the characteristics of the desalination cycle, built on the basis of thermosensitive polyelectrolyte hydrogels, by analytical means. The calculations show that it is possible to select the characteristics of this cycle, which provides a decrease in the concentration of the initial solution of the low molecular target by several times, which, in particular, already ensures the possibility of introducing the water resources available in the vicinity of Astana into economic circulation.

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ПОЛИЭЛЕКТРОЛИТТИ ГЕЛЬДЕРДІҢ КОНТАКТІЛЕНГЕН КЕЗДЕ КОНЦЕНТРАЦИЯНЫ ҚАЙТА БӨЛІНУІ СУДЫ ТҰЩЫЛАНДЫРУДЫҢ ЖАҢА ТЕХНОЛОГИЯСЫ

Аннотация. Аз молекулалы су ерітінділерімен термосезгіш гелдерді суды деминерализациялау үшін қолдануға болатын мүмкіндік көрсетілген. Сондай-ақ ұсынылған әдіс тұзды жерасты суларға, яғни ҚР-ның көптеген аймақтарындағы су тұщылау мәселесін шешуге арналған амал. Мақалада термосезгіш гелдерді қолданысқа енгізу арқылы күн энергиясының радиациясын деминерализацияланған суды алу мақсатында қолдануға мүмкіндік береді. Берілген әдіс концентрацияны қайта құру әсеріне негізделеді, яғни күшті зарядталған гель тұзды суда ісіну кезінде азмолекулярлы компоненті жағынан кедейленген суды сіңіруі арқылы жүзеге асырылады. Тұщылау никлының келесі кезеңінде жиналған су полимерлі тормен жанасып полиэлектrolитті гелден айырылады. Соңында тұщыланған сұйықтық термосезгіш гелді күн сәулесімен қыздыру арқылы алынады. Аралық кезеңді қолдану полиэлектrolитті гелді тікелей қыздыру оның керекті мөлшерде сығуын қамтамасыз ете алмайды, ал термосезгіш гелдердің өзі тор зарядты тығыздыққа ие емес. Мұндай әсерді жүзеге асыру үшін инженерлік есептеулер әдісін ойластыру қажет, және ол термосезгіш гелдің арнайы диапазонды концентрацияны қолдануға болатындай сипаттамаларды анықтауға мүмкіндік беруі тиіс. Бұл жағдай, өз кезегінде алғашқы концентрацияны қайта құру әсерінің теориясын әртүрлі деңгейдегі ионизацияланған екі тордың контактілену жағдайы орын алған кезде жалпылауды талап етеді. Осындай теория осы жұмыста ұсынылған. Оның негізін азмолекулярлы иондардың екі тормен контактіленген кездегі балансын сипаттайтын теңдеулер шешуімен айқындалады. Осындай типті теңдеулер практикалық маңызды жағдайларда аналитикалық шешімдерді талап ететіні іс жүзінде анықталған. Берілген шешімдер жүйе беталысының сапалы түрде сараптамалауына жауап береді. Ұқсату қағидасы орындалады, яғни шектеулі жағдайда алынған нәтижелер бір тордың ионизациялану дәрежесі нөлдік көрсеткішіне ие болғанда, алғашқы ұсынылған теориямен сипатталады. Бұл жағдай берілген теорияны тұщылау никлында қолданылатын термосезгіш гелдердің сипаттамаларын инженерлік есептеу негізінде анықтау үшін қамтамасыз етеді.

Түйін сөздер: су ерітіндісінің деминерализациялануы, термосезгіш гель, су тұщылау мәселелері, аз молекулалы компонент, тұзды су.

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ПЕРЕРАСПРЕДЕЛЕНИЕ КОНЦЕНТРАЦИЙ ПРИ КОНТАКТЕ ПОЛИЭЛЕКТРОЛИТНЫХ ГИДРОГЕЛЕЙ КАК ОСНОВА НОВЫХ ОПРЕСНИТЕЛЬНЫХ ТЕХНОЛОГИЙ

Аннотация. Продемонстрирована возможность использования термочувствительных гидрогелей для деминерализации водных растворов низкомолекулярных солей. В частности, предложенный способ применим к засоленным грунтовым водам, проблема опреснения которых остро стоит для многих регионов РК. Показано, что использование термочувствительных гидрогелей позволяет перейти к использованию энергии солнечной радиации для получения деминерализованной воды. Предложенный способ основывается на эффекте перераспределения концентраций, за счет которого сильно заряженный гель при набухании в солевом растворе поглощает обедненную по низкомолекулярной компоненте воду. На следующей стадии опреснительного цикла аккумулированная жидкость отделяется от полиэлектролитного геля при его контакте с термочувствительной полимерной сеткой. На завершающей стадии опресненная жидкость отделяется от термочувствительного геля за счет нагрева солнечными лучами. Необходимость использования промежуточной стадии определяется тем, что прямой нагрев полиэлектролитного геля не обеспечивает его коллапс, а термочувствительные гели сами по себе не обладают плотностью сеточного заряда, достаточной для выраженного проявления эффекта перераспределения концентраций. Для реализации рассматриваемого способа опреснения необходимо создание методики инженерного расчета, позволяющего определить характеристики термочувствительного геля, используемого для конкретного диапазона концентраций. Это, в свою очередь, требует обобщения ранее развитой теории эффекта перераспределения концентраций на случай контакта двух сеток с неодинаковой степенью ионизации. Такая теория предложена в данной работе. Ее основой является решение уравнений, описывающих баланс концентраций низкомолекулярных ионов при контакте двух сеток. Установлено, что в важных для практики случаях уравнения данного типа допускают аналитические решения. Данные решения отвечают качественному анализу поведения системы. Выполняется принцип соответствия, т.е. полученные решения в предельном случае нулевой степени ионизации одной из сеток переходят в результаты ранее развитой теории. Это обеспечивает использование предложенной теории в качестве основы для инженерного расчета характеристик термочувствительных гидрогелей, используемых в опреснительном цикле.

Ключевые слова: деминерализация водных растворов, термочувствительный гель, проблемы опреснения, низкомолекулярный компонент, соленые воды.