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INFLUENCE THE TEMPERATURE AND CONCENTRATION OF LEAD (II) ON ITS SORPTION BY ACIDOMODIFIED ZEOLITE FROM CONCENTRATED PHOSPHORIC ACID

Abstract. The sorption of lead (II) cations from concentrated phosphoric acid (85.2 %) was studied by the acid-modified zeolite of the Shankanay deposit. The mutual influence of temperature and concentration of Pb^{2+} cations on the process of their sorption is revealed, the time has no significant effect.

The sorption curves of the dependence of the residual content of lead (II) on the temperature in phosphoric acid with a content of 10 and 55 mg/Pb have a minimum at 43 °C, corresponding to its maximum absorption. In phosphoric acid with C_{Pb} equal to 28 mg/l with increasing temperature, the sorption curves increase, and with C_{Pb} equal to 100 mg/l - decrease, respectively, the degree of sorption decreases and increases. The curves for the residual content of lead (II), depending on its concentration up to 30 °C, have a minimum at C_{Pb} of 28 mg/l, corresponding to a high degree of lead sorption (99.0-99.6 %), and in the range from 45 to 70 °C - a maximum characterizing the lower degree of Pb absorption. Moreover, the maximum on the sorption curves with increasing temperature shifts to a region of solutions less concentrated in lead content. The appearance of a maximum or minimum on the sorption curves of Pb^{2+} cations is due to the desorption process, when under the conditions under study the sorbed cations from the zeolite exit to the solution.

Keywords: heavy metals, lead cations, acid-modified zeolite, sorption, phosphoric acid.

Introduction. Studies of the adsorption properties of natural zeolites to a number of heavy metals in aqueous media have been widely performed in [1-4]. Structural features of natural zeolites determine the penetration of not only metal cations with large radii, but also molecules into the crystalline structure of natural zeolite [1, 5]. During sorption, the ions (molecules) of the sorbate migrate from the volume of the medium to the surface of each body and pass into its adsorption layer. Adsorbed ions migrate in this layer, diffuse into the body volume or are desorbed. If the interaction of sorbate molecules with each other is sufficiently strong, they are assembled into two-dimensional clusters that can expand along the surface until they come into contact with neighboring clusters [6]. Migrating along the surface, the molecules for some time are in some adsorption center, and then jump to the next center. In this case, the residence time of a molecule on the vertices, edges or steps on the faces of a single crystal can differ by a factor of tens. The difference in the residence of molecules in adsorption centers located on crystalline faces with different atomic relief is equally great. Therefore, faces with

an unequal number of steps have different sorption capacity: each face is characterized by its own local adsorption coefficient and sorption capacity.

In recent years, sorption purification methods using activated zeolites have become widespread [7, 8]. The acid activation of natural zeolites is accompanied by a process of dealumination, which is caused by the unblocking of the channels of the zeolite framework, which leads to an increase in the Si/Al ratio and the formation of silanol groups, an increase in the effective size of micropores, and an increase in the capacity of zeolite exchange [9-13]. Along with the appearance of active in the sorption of H^+ exchange acid sites and the displacement of aluminum (Al^{3+}) in the exchange positions, as a result of acid activation, the arrangement of active sites is more favorable for the interaction of reacting substances. In addition, when acidizing zeolite, the liberated silica increases the specific surface area and porosity of the activated samples and removes impurities blocking the channels. The stability of the silica-alumina skeleton of high-silica zeolites to the action of acids has increased the possibilities of regulating their properties by decationizing and dealumination under various conditions of acid treatment.

A significant number of works are devoted to IR spectroscopic investigation of clinoptilolite after hydrochloric acid activation [14-19]. According to the obtained data, silanol groups Si-OH, bound by hydrogen bonds, removal of aluminum from the sorbent framework, and formation of "hydroxyl nests" associated with the formation of Al-O vacancies occur in the structure of the natural zeolite after acid treatment. In this case, an increase in the amount of loosely bound water and a decrease in water molecules involved in the formation of hydrogen bonds with silanol groups and the hydration of out-of-frame cations occur.

It has been found that acid-activated clinoptilolite tuffs exhibit high sorption properties, for example with respect to phenylalanine [20], formaldehyde [21], NH_4^+ ions [22], and SO_2 molecules [23]. Acid modification of natural zeolite allows to obtain efficient sorbents with respect to cations of heavy metals in aqueous media [24, 25]. It can be seen from the review that the applications of zeolites in various industries can be significantly expanded by modifying them. The aim of this work was to study the sorption of Pb (II) cations by the acid-modified zeolite from the Shankanay deposit from concentrated phosphoric acid.

Experimental part

Lead-containing phosphoric acid with a predetermined concentration of Pb^{2+} cations was prepared by introducing in the H_3PO_4 (85.2 %) the calculated amount of lead acetate $Pb(CH_3COO)_2 \cdot 3H_2O$ of the grade "chemical pure". As a sorbent, the zincite of the Shankanay deposit, modified with 15 % hydrochloric acid, was used. The sorption of lead (II) cations was carried out in a thermostated reactor under the conditions of mixing the sorbent (Solid) with lead-containing phosphoric acid (Liquid), taken in a ratio of S:L equal to 10:100. The initial and final concentrations of lead ions in solutions were determined on an atomic absorption

spectrophotometer type AA-7000, the company "Shimadzu Corporation", Japan, No. A30664901456. The relative standard deviation of the output signal is 2 %.

Sorption was assessed by the change in the content of Pb^{2+} cations in solution, i.e. by the difference between the initial concentration and its residual concentration. The degree of sorption (extraction) is the ratio of the difference between the initial and reached the concentration of the cation Pb (II) to its original content reached at a fixed time.

The degree of purification of various media, including phosphoric acid, depends significantly on the main parameters of the sorption process (temperature, cation concentration Pb^{2+} , time). A study of the mutual influence of these technological factors on the process of purification of phosphoric acid from lead ions ((II)) with the acid-modified zeolite of the Shankanay deposit was carried out using the method of mathematical planning of the orthogonal rotatable 3 factor experiment of the 2nd order [26].

Variable (input) factors of the process were: Z_2 - temperature (20 -70°C); Z_3 - C concentration of cations Pb^{2+} (10-100 mg / l); Z_1 - time (5 - 60 minutes). These changes in the concentration of Pb (II) cations correspond to a possible range of their presence in acidic effluents and other contaminated solutions, as well as in thermal and extraction phosphoric acid.

The determined (output) parameter Y1 (response) was the residual content of Pb^{2+} ions in the liquid phase of the " Pb^{2+} acid acid-acid-modified zeolite" system under investigation after its purification by the sorbent.

The coordinates of the center of the plan, the variation intervals and the study levels are represented by the natural values Z_1, Z_2, Z_3 in table 1.

Table 1 – Coordinates of the center of the plan, levels of variation

Value of variables			
Coded	Natural		
	Z_1 , time, min	Z_2 , T°C	Z_3 , mg / l Pb^{2+}
The upper level (+1)	48,9	59,9	81,8
The center of the plan is the zero level (Z_i^0)	32,5	45	55
The lower level (-1)	16,1	30,1	28,2
Interval of variation along the axis (ΔZ_i)	16,4	14,9	26,8
Star shoulder (+1,682)	60	70	100
Star shoulder (-1,682)	5	20	10

The experimental conditions and the results obtained in table 2.

With the use of computer processing of experimental data, a regression analysis of the results was carried out. A regression equation describing the dependence of the residual lead content in an acid medium after its removal by an acid-modified zeolite on the process parameters under study was obtained by correlation analysis after elimination of insignificant coefficients.

Table 2 – Process conditions, content of Pb²⁺ sorption cations and the degree of their sorption by acid-modified zeolites in acid medium

No. of experience	The intrinsic value of experiments			Content of Pb ²⁺ after sorption, mg/l	The degree of sorption (R), %
	X ₁ , min	X ₂ , °C	X ₃ , mg/l		
1	16,1	30,1	28,2	4,03	85,71
2	48,9	30,1	28,2	4,13	85,35
3	16,1	59,9	28,2	4,42	84,32
4	48,9	59,9	28,2	5,21	81,52
5	16,1	30,1	83,7	9,36	88,81
6	48,9	30,1	83,7	9,42	88,74
7	16,1	59,1	83,7	9,46	88,69
8	48,9	59,1	83,7	1,05	98,74
9	5	45	55	5,78	89,49
10	60	45	55	5,94	89,20
11	32,5	20	55	6,41	88,34
12	32,5	70	55	7,74	85,92
13	32,5	45	10	1,16	88,40
14	32,5	45	100	1,04	98,96
15	32,5	45	55	5,44	90,11
16	32,5	45	55	5,69	89,65
17	32,5	45	55	3,20	94,18
18	32,5	45	55	5,82	89,41
19	32,5	45	55	7,14	87,01
20	32,5	45	55	6,51	88,16

Results and discussion

After processing the obtained data and excluding the insignificant factors, the regression equation is given below, in which six coefficients are significant:

$$Y=5,793-0,527X_1+0,493X_2+1,805X_3-1,326X_3^2-2,238X_2X_3 \quad (1)$$

The adequacy of the regression equation was verified by the Fisher criterion. As a result of optimization of the calculations, it was found that the calculated value of the Fisher test is less than the tabulated value:

$$F < F_{1-p} \quad (2)$$

namely 4.2728734016 < 4,69999998, therefore, the equation adequately describes the experiment.

An analysis of the regression equation obtained showed that the temperature and concentration of Pb²⁺ ions have the greatest influence on the sorption of

Pb (II) cations. Comparison of the coefficient values showed that the time has no significant effect on the output parameter.

As already noted, temperature has a significant effect on sorption purification processes, using both natural aluminosilicate sorbents and their modified forms as sorbents. According to the regression equation 1 describing the sorption of Pb^{2+} cations by an acid-modified zeolite in phosphoric acid, the effect of temperature on the output parameter is complex and is represented by positive linear (X_2) and double interaction of process temperature and lead cation (X_2X_3) concentration with a minus sign. In the regression equation, before the variable temperature coefficient (X_2) there is a plus sign. That is, the temperature factor affects the output parameter not only as a double interaction with the third factor (X_2X_3), but also independently, and directly proportional. A comparison of the values of the coefficients showed that the greatest influence on the output parameter is exerted by a double interaction of the process temperature and the concentration of Pb^{2+} cations. It follows from the regression equation 1 that the double interaction (X_2X_3) has a minus sign at a coefficient. That is, a decrease in the value of only one factor (either temperature or Pb concentration) can increase the purity of phosphoric acid. The simultaneous decrease or increase in the values of both factors in the regression equation when they multiply gives a plus sign and, as a result, a decrease in the degree of purification. Simultaneous decrease in the values of temperature and concentration leads to poor purification of the acid medium, and a simultaneous increase in their values may contribute to the desorption (exit) of lead ions from the sorbent solution.

The graphical dependence of the obtained dependences of the residual content of Pb^{2+} ions in the liquid phase of the " Pb^{2+} - H_3PO_4 -acid-modified zeolite" system on temperature is shown in Fig. 1. It can be seen that in low- and medium-concentrated lead-containing solutions (10 and 55 mg / l Pb) cations of Pb^{2+} are extreme with a minimum at 45°C, which is most pronounced in solutions obtained with C_{Pb} equal to 10 mg/l, and corresponds to the highest degree of sorption of Pb^{2+} cations (figure 1a, c). For example, 47.5 % Pb is sorbed from an acidic solution with C_{Pb} equal to 10 mg / l at 20 ° C for 16 minutes, 99.5 % Pb at 45 ° C, 63 % Pb at 60 ° C, and Pb at 60°C, 28 % Pb. Moreover, in low-concentration solutions of lead, an increase in the duration of the process has a slight negative effect on the sorption capacity of the sorbent. That is, with decreasing phase contact time, the degree of sorption somewhat increases as the temperature decreases. Thus, 99.5 % Pb sorbed in 5 min at 45 ° C, 95 % Pb in 16 min, 91 % Pb in 32 min, 84 % Pb in 50 min and 81 % in 60 min, 3 % Pb.

In solutions with a lead content of 28 mg/l, the extreme dependence of the sorption curves on temperature is smoothed (figure 1b), i.e. As the temperature rises, the residual content of Pb (II) cations constantly increases, so the degree of sorption decreases. Thus, 95.7 % Pb is sorbed in this solution for 5-16 min at 20 °C, 94.04 % Pb at 30 °C, and 69.85 % Pb for 60 min.

In solutions with a lead concentration of 82-100 mg/l, an increase in the process temperature has a positive effect. Under the investigated conditions, the

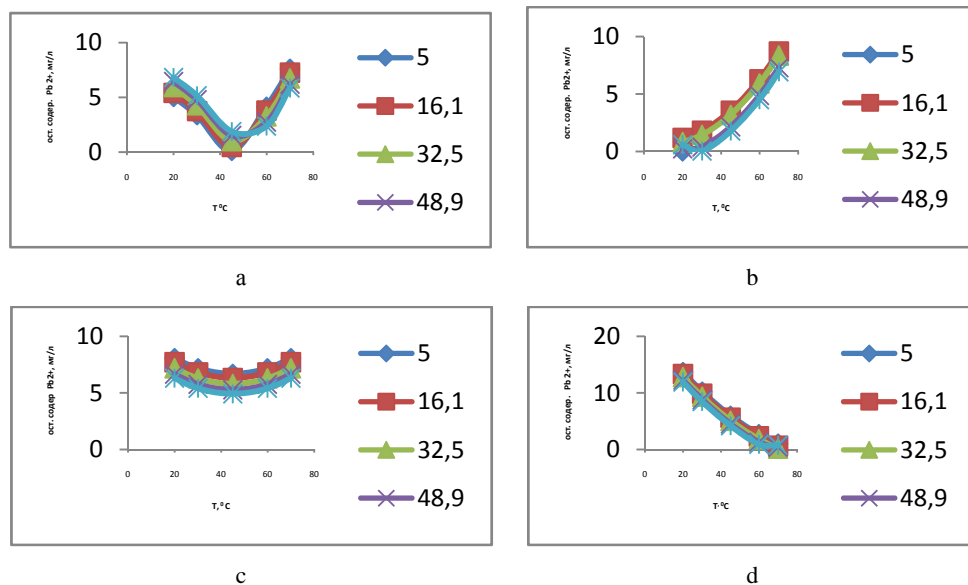
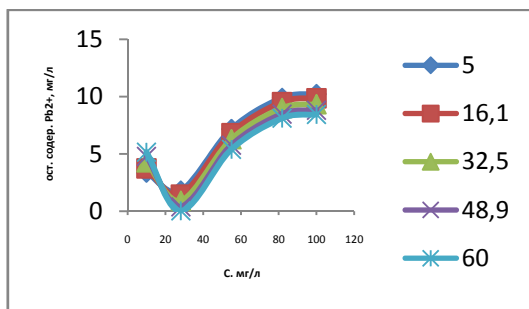


Figure 1 – Effect of temperature on the residual content of Pb (II) cations in the system «Pb²⁺ - H₃PO₄ - acid-modified zeolite»: a – 10 mg/l Pb²⁺; b – 28 mg/l Pb²⁺; c – 55 mg/l Pb²⁺; d – 100 mg/l Pb²⁺

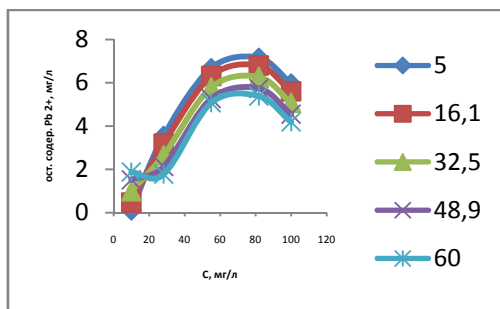
residual content of Pb²⁺ cations in the liquid phase of the system decreases with increasing temperature, and the degree of their sorption increases accordingly (figure 1d). For example, in a solution with C_{Pb} equal to 100 mg/l at 20°C and 16 min, the sorption degree of Pb²⁺ ions reaches 86.9%, at 40 °C - 94.2%, at 70 °C - 99.1 %.

The effect of the concentration of Pb²⁺ cations on its residual content in the system under study is complex and is represented in the regression equation 1 by a positive linear concentration factor of lead (X₃) and its negative quadratic term (X₃²). In this case, the values of the coefficients of the factor under consideration are C_{Pb} (X₃) higher than in X₁ (time), which indicates a significant influence of lead concentration on the process of its sorption compared with time.

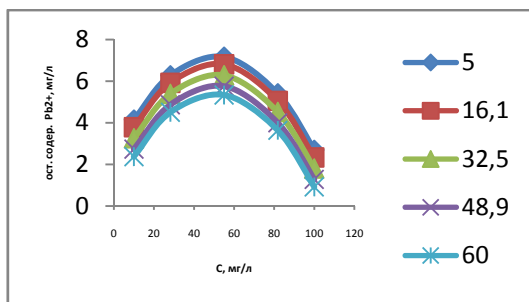
It should be taken into account that in the regression equation 1 the influence of C_{Pb} is also represented by a negative double factor of its interaction with the process temperature (X₂X₃). A comparison of the values of the X₂X₃ coefficients showed that a double interaction of the Pb²⁺ cation concentration and the process temperature has a significant effect on the output parameter. The minus sign at the coefficient of variables X₂X₃ in the regression equation shows that the simultaneous increase or decrease of the values of these factors will not lead to the purification of the acidic environment. On the contrary, a decrease in the value of one of the factors with an increase in the value of the other leads to an increase in the degree of sorption of lead. The considered interference of factors and their effect on the response is well demonstrated by the graphs below (figure 2). In the



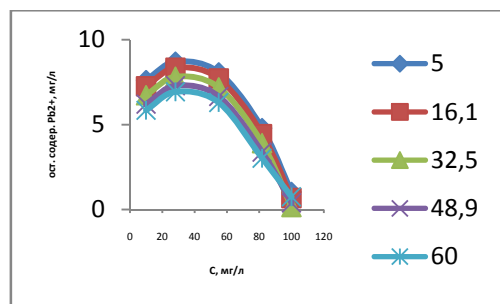
a



b



c



d

Figure 2 – Influence of the concentration of cations of Pb (II) on its residual content in the system «Pb²⁺ - acidic medium - acid-modified zeolite»: a – 30 °C; b – 45 °C; c – 60 °C; d – 70 °C

regression equation 1, the plus sign is used before the coefficient of variable lead concentration (X_3). That is, the concentration factor of lead (II) affects the output parameter not only in the form of negative quadratic and double interaction with the second factor (temperature), but also independently, and directly proportional. The higher the concentration of lead in solution, the more it remains after the end of the sorption process. At the same time, the value of the coefficient X_3 shows a greater effect than the influence of its quadratic factor X_3^2 , but a slightly smaller effect compared to the double factor X_2X_3 . That is, even in lead-concentrated solutions, the acid-modified zeolite will exhibit rather high sorption properties.

The curves of the residual content of Pb^{2+} ions as a function of their concentration over the whole time interval studied at 20-30⁰C have a minimum in phosphoric acid with C_{Pb} equal to 28 mg/l, which corresponds to the maximum absorption of Pb^{2+} cations by acid-modified zeolite 99.0-99.9 % (figure 2 a).

The sorption curves of lead depending on its concentration at elevated temperatures from 45 to 70⁰C are characterized by a clearly pronounced maximum, which is due to the influence of the double factor X_2X_3 (figure 2b, c, d). The sorption capacity of the acid-modified zeolite under these conditions increases slightly with the increase in the concentration of Pb^{2+} cations, and then increases. For example, the degree of sorption in a system with C_{Pb} equal to 10 mg/l reaches 90 %, with C_{Pb} equal to 40 mg/l – 86 %, C_{Pb} equal to 73.5 mg/l - 91.4 %, and with C_{Pb} equal to 100 mg/l -94.8 % at 45 ⁰C for 16 minutes. Moreover, the maximum on the sorption curves shifts to the region of lower concentrations with increasing temperature. So the maximum is prescribed in H_3PO_4 with a concentration of 73.5 mg/l Pb^{2+} at 45 ⁰C, with C_{Pb} equal to 52.6 mg/l Pb^{2+} at 60 ⁰C, and with C_{Pb} equal to 31.6 mg /l Pb^{2+} at 70⁰C. At the same time, a high degree of sorption of 99.0-99.8 % is already achieved in phosphoric acid with C_{Pb} equal to 100 mg/l. It is possible that at an elevated temperature in concentrated phosphoric acid the process of dealumination continues, which causes an increase in the sorption capacity of the acid-modified zeolite with phosphoric acid with a high content of Pb^{2+} cations. That is, in the purification of lead-containing phosphoric acid by acid-modified zeolite, the concentration of lead must be taken into account. A high degree of purification can be achieved practically in the entire investigated range of Pb (II) ion concentrations, selecting the process temperature.

It should be noted that the presence of a maximum or minimum on the sorption curves appears to be due to the saturation of the zeolite and, as a consequence, the deterioration of its sorption properties. This process, in turn, may be caused by the system's striving for equilibrium, in which the concentration of lead in both phases is equalized. That is, the appearance of a maximum on the sorption curves is due to the desorption process, when under these conditions the sorbed cations from the zeolite exit to the solution.

Thus, when choosing the optimal conditions for the purification of concentrated phosphoric acid from cations of Pb^{2+} with an acid-modified zeolite, the temperature of the process and the concentration of lead (II) cations should be taken into account first. On the basis of the studies, optimum conditions for the

maximum hollow purity of a low-concentration lead (10 mg/l Pb) of phosphoric acid were found: time - 16 min, temperature – 45 °C; more concentrated in lead (28-100 mg/l Pb) of H₃PO₄, the time is 50 minutes, and the temperature is 40 °C, and in high-concentrated for lead H₃PO₄ the temperature needs to be raised to 70 °C.

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Резюме

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ҚЫШҚЫЛМЕН МОДИФИКАЦИЯЛАНҒАН ЦЕОЛИТ АРҚЫЛЫ КОНЦЕНТРЛІ ФОСФОР ҚЫШҚЫЛЫНАН ҚОРҒАСЫННЫҢ СОРБЦИЯЛАНУЫНА ТЕМПЕРАТУРА МЕН ҚОРҒАСЫН (II) КОНЦЕНТРАЦИЯСЫНЫҢ ӘСЕРІ

Шаңқанай кен орнының қышқылмен модификацияланған цеолиті арқылы концентрлі фосфор қышқылынан (85,2 %) қорғасын (II) катиондарын сорбциялау процесі зерттелді. Температура мен Pb²⁺ катиондары концентрациясының олардың сорбциялану процесіне өзара әсер ететіні анықталды, ал уақыт онша әсер етпейді.

Құрамында 10 және 55 мг/Рb бар фосфор қышқылындағы қорғасынның (II) қалған мөлшерінің температураға тәуелді сорбциялық қисықтарының 43°C-та минимумы бар, яғни бұл оның ең көп мөлшерде сорбцияланғанын көрсетеді. Құрамында 28 мг/л С_{Рb} бар фосфор қышқылында температура артқан сайын сорбция қисықтары жоғарылайды, ал құрамында 100 мг/л С_{Рb} бар фосфор қышқылында төмендейді, яғни сәйкесінше сорбция дәрежесі азаяды және артады. Қорғасынның (II) концентрацияға тәуелді қалған мөлшері 30 °С С_{Рb} концентрациясы 28 мг/л тең болғанда минимумы бар, ол қорғасынның жоғарғы сорбциялану дәрежесіне (99,0-99,6 %) сәйкес келеді, ал 45-тен 70 °С-қа дейінгі температура аралығында қорғасынның төменгі сорбциялану дәрежесін көрсететін максимум бар. Сорбциялық қисықтарда температура артқан сайын пайда болатын максимум қорғасын мөлшері бойынша аз

концентрі ерітінділер жаққа қарай ығысады. Pb^{2+} катиондарының сорбциялану қисықтарында максимумның немесе минимумның пайда болуы десорбциялану процесімен түсіндіріледі, яғни зерттеліп отырған жағдайларда сорбцияланған катиондардың цеолиттен қайтадан ерітіндіге өтуі орын алады.

Түйін сөздер: ауыр металдар, қорғасын катиондары, қышқылмен модификацияланған цеолит, сорбция, фосфор қышқылы.

Резюме

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ВЛИЯНИЕ ТЕМПЕРАТУРЫ И КОНЦЕНТРАЦИИ СВИНЦА (II) НА ЕГО СОРБЦИЮ КИСЛОТОМОДИФИЦИРОВАННЫМ ЦЕОЛИТОМ ИЗ КОНЦЕНТРИРОВАННОЙ ФОСФОРНОЙ КИСЛОТЫ

Исследован процесс сорбции катионов свинца (II) из концентрированной фосфорной кислоты (85,2%) кислотомодифицированным цеолитом месторождения Шанканай. Выявлено взаимовлияние температуры и концентрации катионов Pb^{2+} на процесс их сорбции, время не оказывает существенного влияния.

Сорбционные кривые зависимости остаточного содержания свинца (II) от температуры в фосфорной кислоте с содержанием 10 и 55 мг/ Pb имеют минимум при 43°C, соответствующий его максимальному поглощению. в фосфорной кислоте с C_{Pb} равной 28 мг/л с повышением температуры кривые сорбции возрастают, а с C_{Pb} равной 100 мг/л – снижаются, соответственно степень сорбции уменьшается и увеличивается. Кривые остаточного содержания свинца (II) в зависимости от его концентрации до 30°C имеют минимум при C_{Pb} равной 28 мг/л, соответствующий высокой степени сорбции свинца (99,0-99,6 %), а в области от 45 до 70°C - максимум, характеризующий более низкую степень поглощения Pb. Причем максимум на сорбционных кривых с ростом температуры смещается в область менее концентрированных по содержанию свинца растворов. Появление максимума либо минимума на кривых сорбции катионов Pb^{2+} обусловлено процессом десорбции, когда в исследуемых условиях происходит выход сорбировавшихся катионов из цеолита обратно в раствор.

Ключевые слова: тяжелые металлы, катионы свинца, кислотомодифицированный цеолит, сорбция, фосфорная кислота.