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Efficiency of industrial wastewater treatment using natural bentonite sorbents

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Abstract. The article is devoted to the current problem of the industrial town Tekeli of the Zhetysu Region, the Republic of Kazakhstan, in the area of operation the Tekeli mining and processing complex, based on TMPC LLP, where widespread pollution of water resources is observed. The main pollutant of its water basin is industrial waste of the operating enterprise located on the sites of the mining and processing complex. The water of the Karatal River is under intense pressure from the toxic components of the mining industry, which is located in the zone of the densest river network. This co-arrangement contributes to the fact that substances with gaseous, liquid and solid waste inevitably enter the river network. As a result, the nature of the deterioration in surface water quality in the Zhetysu region is becoming a steady trend. Of significant interest for the development of sorption methods for the purification of industrial effluents is the use of aluminosilicates - bentonites, as the most common and cheap. However, in a natural state without activation, they often do not have a high sorption capacity, which entails an increased consumption of them. It became necessary to obtain activated sorbents with a higher sorption capacity from natural mineral raw materials of aluminosilicates. The purpose of this work is to develop an innovative technology for cleaning industrial effluents from heavy metals using a natural sorbent – bentonite. The authors in the study used bentonite as a sorbent, as the most common and cheapest sorbent in this region. However, in their natural state without activation they often do not have a high sorption capacity, which causes increased consumption. There was a need to obtain activated sorbents with a higher sorption capacity from the natural mineral raw material bentonite for development technologies of advanced treatment of industrial effluents containing heavy metals (Zn^{2+}, Pb^{2+}, Cu^{2+}).

Keywords: *aluminosilicates-bentonites, sorption capacity, industrial effluents of the enterprise, sorption of heavy metals, toxicants, Karatal river.*

1. Introduction

Relevance of the work. Currently, the treatment of industrial effluents of the TMPC LLP enterprise is an urgent environmental problem in this region. Despite all the measures and methods used to treat the effluents, pollutants continue to enter the water bodies. The most dangerous pollutants are heavy metals (HM).

Adsorption methods are the simplest, least expensive, accessible and effective of all the special methods of water treatment. In the Zhetysu Region there are deposits of bentonites, which are used as sorbents for advanced treatment of industrial effluents. Bentonites have increased selectivity to heavy metal ions and polar substances, which allows us to expect high efficiency in industrial effluents treatment processes [1-3]. Bentonite is a complex mineral, whose composition is determined by the content of montmorillonite in clay, having the formula $Si_8\ Al_4\ O_{20}\ (OH)_4 \times nH_2O$, where silicon can be replaced by various cations (aluminum, iron, zinc, magnesium, calcium, sodium, potassium, etc.) [1, 3-4].

The region's water resources are under intense pressure from toxic components of industrial mining enterprises located in the area of the densest river network. This co-location leads to the fact that substances with gaseous, liquid and solid waste inevitably end up in the river network. As a result, deterioration in the quality of surface waters in the Zhetysu region is becoming a stable trend. The Karatal River is one of the most polluted rivers of the Republic of Kazakhstan, and the highest pollution of surface waters is observed in the area of Tekeli mining and processing complex. According to the RSE «Kazhydromet» [1, 5-6], the most polluted river in terms of chemical indicators is the Karatal River. The pollution index of this river has remained high over the past 20 years.

Based on the analysis of the results of long-term experimental studies of water supply and sanitation systems of the industrical cleaning filters, carried out by Evdokimov P.D., Sazonov G.T., Berger B.S., Bagrov O.N., Antonov V.N., Myrzakhmetov M.M., Golovanov G.A., scientific and technical principles and methodological foundations for creating low-waste water supply and industrial wastewater disposal systems of mining and processing complex have been developed [1, 2].

Among the known environmental pollutants, the most dangerous toxicants are heavy metals that have the ability to propagate in gaseous, liquid and solid phases. The study found out that most often in the industrial wastewater of the Tekeli mining and processing complex there are the follow-

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ing metals: Pb^{2+}, Zn^{2+}, Cu^{2+}, Cd^{2+}, Fe^{2+}, Cr^{6+}, Mn^{2+}, Al^{3+} in mobile water-soluble forms, as well as in the form of suspension [3, 7-8].

Scientific work is devoted to development of industrial wastewater treatment technology of Tekeli mining and processing complex with natural sorbents, bentonites of the Mukry deposit in the Zhetysu Region of the Republic of Kazakhstan. The research work was carried out in accordance with the research plan of the scientific and research work carried out over a number of years under the educational program «Natural and Technical Sciences» of Zhetysu University named after I. Shinzaburo.

The significant interest to us was the use of sorption method of purifying industrial wastewater with aluminosilicates-bentonites, as the most common and cheapest. However, in their natural state without activation, they often do not have a high sorption capacity, which entails increased consumption [9, 10]. There was a need to obtain activated sorbents with a higher sorption capacity from the natural mineral raw material, aluminosilicate-bentonite, for the further development of technologies for purification of industrial wastewater containing heavy metals (Zn^{2+}, Pb^{2+}, Cu^{2+}, etc.).

The purpose of this work is to develop an innovative technology for purifying industrial wastewater from heavy metals using a natural sorbent, bentonite.

Monitoring of the surface water ecosystem of the studied area and [11] in particular the river Karatal along its entire length shows that water ecosystem is moderately polluted.

Intense and long-lasting zones of pollution have formed in the bottom sediments of rivers, which are secondary sources of pollution of the reservoir.

Pollution of the surface reservoir in the areas where the Tekeli mining and processing complex operates has led to an unacceptable deterioration of the quality of groundwater. Natural purification when filtering through the soil does not give the desired effect, since the filtering soil rocks themselves are saturated with chemical compounds, as a result of which the concentration of heavy metals in groundwater increases.

The liquid phase of industrial wastewater in the tailing storage contains a significant amount of free oxygen and CO_3 than groundwater [12-13], which creates favorable conditions for intensive desolination from industrial wastewater with transfer into the liquid phase of a significant number of chemical elements and their compounds that pollute the environment.

2. Materials and methods

Environmental hazards of industrial waste of Tekeli mining and processing complex for groundwater and surface water was assessed by chemical indicators of surface water according to the method [14-15] proposed for calculating the water pollution index (WPI).

Water quality was assessed by WPI using formula (1):

$$WPI = \frac{C / MPC}{n}, \quad (1)$$

where C is the actual concentration of the pollutant;

MPC – maximum permissible concentration of a pollutant;

n – the number of the most dangerous pollutants.

The quality of the surface water on hydrochemical indicators of the WPI of the RSE «Kazhydromet» of Taldykorgan city is shown in Table 1.

Table 1. Water quality class, based on the calculated WPI value

WPI value	Water quality class	Nature of quality
<0.3	1	very clean
0.3-1.0	2	clean
1.0-2.5	3	moderately polluted
2.5-4.0	4	polluted
4.0-6.0	5	dirty
6.0-10.0	6	very dirty
>10.0	7	extremely dirty

3. Results and discussion

Analysis of WPI indicator made it possible to compare the water quality of different objects, regardless of the presence of various pollutants, and to identify the dynamics of changes in water quality over the years. The Kora River is the inflowing stream of the Karatal River in the area where the Tekeli mining and processing complex is located. Industrial waste of the Tekeli factory is contaminated with copper, zinc, lead ions, as well as sulfates. Industrial waste of the mining and processing complex, drainage waters from tailing storages, waste and mine waters pollute the groundwater that feeds the Kora River. As evidenced by the high content of heavy metals (Cu^{2+}, Pb^{2+}, Zn^{2+}, Cd^{2+}) in the Kora River, given in Table 2.

Table 2. Content of heavy metals (Cu^{2+}, Pb^{2+}, Zn^{2+}, Cd^{2+}) in the Kora River in the area of industrial cleaning filters operation

Heavy metals	Maximum value, mg/dm^3	Average value, mg/dm^3	Minimum value, mg/dm^3	Exceeding the maximum permissible concentration $_{px}$*, times		
				max. values	average value	minimum values
Cu^{2+}	1.73400	0.017	0.00620	173.4	17.33	6.2
Pb^{2+}	1.5600	0.009	0.00050	15.6	0.09	0.005
Zn^{2+}	1.4200	1.146	0.00050	142	114.6	0.05
Cd^{2+}	0.00900	0.006	0.00008	1.8	1.2	0.02

*Note: according to Sanitary Regulations and Standards of the Republic of Kazakhstan 3.01.067.97, the maximum permissible concentration $_{px}$ is 0.001 mg/dm^3 for copper, 0.01 mg/dm^3 for zinc, 0.1 mg/dm^3 for lead and 0.005 mg/dm^3 for cadmium.

The maximum excess of the maximum permissible concentration $_{px}$ for copper is 173.4 times and for zinc 142 times, which occurs due to the high content of copper and zinc in the factory water discharged into the Kora River without treatment.

In 2022, the water condition of the Kora River was assessed as «extremely dirty»; the WPI is 14.94 according to Table 1 and exceeds the norm by 7.5 times.

For industrial wastewater treatment of the mining and processing complex from heavy metals Cu^{2+}, Pb^{2+}, Zn^{2+}, Cd^{2+} and other pollutants the method of accelerating the process of sedimentation of finely dispersed impurities and emulsified substances, coagulation, is used. As a result of coagulation, flakes of metal hydroxides were formed (with the ability to trap colloidal and suspended particles), which were then deposited under the influence of gravitational forces [16-19, 26].

When suspended particles are aggregated with addition of high molecular weight compounds to industrial wastewater, the process of flocculation occurs. Use of flocculants made it possible to cut down on the dose of coagulants and reduce the duration of coagulation process, as well as to increase the rate of sedimentation of the resulting flakes. To intensify the process of floc formation and increase the rate of their deposition, aluminum and iron hydroxides were used [5, 20-21].

The sorption methods we use allow us to carry out a higher degree of purification of industrial wastewater, up to 80% and higher, compared to chemical reagent methods. These methods are based on the processes of contact sorption of substances on the developed adsorbents surface.

In the studies of Sh.B. Battalova, M.M. Kravchenko [16, 22-23], it was theoretically and experimentally proven that bentonites of the 14th horizon of the Mukry deposit have the greatest sorption capacity, as shown in Table 3, they have a monomineral composition, the content of the mineral montmorillonite reaches 90% [8, 17, 25].

Bentonite of the Mukry field was used in the studies for industrial wastewater treatment of the mining and processing complex from heavy metals Pb^{2+}, Cu^{2+}, Zn^{2+}, Cd^{2+}. A series of experiments were carried out using model solutions of heavy metals, as well as industrial waste drains with thickeners of copper-zinc and lead concentrates of the mining and processing complex. The experiments were carried out under static conditions, at a pH of the environment from 7.5 to 8.5.

In the experiments, bentonite was used without activation at natural humidity 8%, the optimal contact time of the sorbent with wastewater was 2 hours, the mass of the sorbent (g/dm^3) was 20, 30 and 40.

Table 3. Results of industrial wastewater treatment depending on the amount of used sorbent

Name of heavy metal	Sorbent mass, g /dm^3	Initial conc. C_{ref}, g/dm^3	Residual concentration C_{res}, g/dm^3 at contact time, hour				Purification degree α, % at contact time, hour			
			1	1.5	2	2.5	1	1.5	2	2.5
Cu^{2+}	20	5	4.30	3.74	3.49	3.66	17	26	31	28
	30	5	3.72	2.39	2.27	2.48	26	47	56	51
	40	5	3.48	2.58	1.82	1.88	31	53	65	60
Pb^{2+}	20	4.48	3.32	1.33	2.90	3.05	25	32	37	34
	30	4.48	3.28	1.86	2.23	2.44	26	41	51.5	47
	40	4.48	2.75	2.41	1.48	1.77	37	53	64.5	62
Zn^{2+}	20	1.06	0.83	0.30	0.65	0.71	22	31	36	33
	30	1.06	0.76	0.42	0.53	0.62	25	37	45.1	42
	40	1.06	0.70	0.54	0.41	0.46	36	48	58	54

Increasing the contact time of the sorbent and industrial waste by more than 2 hours leads to decrease in the degree of purification, as the desorption process begins. To achieve MPC $_{px}$ standards, the mass of the sorbent must be more than 50-60 g. This is irrational. The purification degree α is calculated using formula (2):

$$\alpha = \left[\left(C_{ref} - C_{res} \right) / C_{res} \right] \cdot 100\% \qquad (2)$$

where C_{ref} is the concentration of the pollutant in the original solution, mg/dm^3;

C_{res} is residual concentration of pollution in solution, mg/dm^3 [4, 24].

To increase the sorption capacity, the experiment was carried out using the «Magnafloc 10» flocculant and aluminosilicate-bentonite.

We used 20 g/dm^3 of bentonite of horizon 14 on a dry matter basis, the contact time of the sorbent, flocculant and industrial waste was 2 hours. The results of a series of experiments are given in Table 3.

Analysis of the results in Table 3 showed the highest degree of purification 62.4% corresponding to the regime when using 0.5 mg/dm^3 flocculant and 20 g sorbent. When the mass of the sorbent increased to 40 g, the degree of purification increased by 2.6%, since the process of simultaneous sorption and flocculation has a physical limitation: flocculants clog pores accessible to heavy metals.

The experiment showed use of bentonite in the amount of 20 g/dm^3 and the «Magnafloc 10» flocculant in the amount of 0.5 mg/dm^3, the degree of purification from zinc is 66%, from copper – 70% and from lead – 72%. Thus, when using a flocculant-sorbent composite, the degree of copper purification is 70% in the mode of 0.5 g/dm^3 flocculant and 20 g of sorbent as at 40 g/ dm^3 of bentonite in natural occurrence.

During the study period, the situation with pollution of the Karatal River with heavy metals (Cu^{2+}, Pb^{2+}, Zn^{2+}, Cd^{2+}) did not change significantly due to the accumulation of heavy metals in bottom sediments of river beds. The level of WPI was assessed as «high», and in some areas – «very high».

Pollution of the Karatal River is also caused by the activities of the Tekeli mining and processing complex. Sewage and drainage water pollution ranges from «very dirty» (WPI equal to 6) to «extremely dirty» pollution (WPI equal to 7). This has led to pollution of the Kora River, a tributary of the Karatal River into which wastewater is discharged, where WPI amounts to «very high» levels of pollution.

The experiment showed use of bentonite in the amount of 20 g/dm^3 and «Magnafloc 10» flocculant in the amount of 0.5 mg/dm^3 improves the degree of purification of zinc by 66%, copper by 70% and lead by 72%.

Thus, when using a flocculant-sorbent composite, the degree of copper purification is 70% in the mode of 0.5 g/dm^3 flocculant and 20 g of sorbent, as with 40 g/dm^3 of bentonite.

4. Conclusions

The study outlines scientifically based technological and environmental developments for industrial wastewater treatment with acid-alkaline activated bentonites, providing the solution to important scientific and practical problems to reduce the negative impact of industrial wastewater on surface water bodies.

The nature of pollution has been systematized and it has been revealed that the main ones are heavy metals - lead, zinc, copper. The pollution assessment was carried out using the water pollution index (WPI).

An environmental and economic assessment of the effectiveness of sorption technology for purification of heavy metals - Zn^{2+}, Pb^{2+}, Cu^{2+} with acid- alkaline activated bentonites is presented.

Based on the results of pilot tests of purification of industrial wastewater from heavy metals with acid-alkali activated bentonites, environmental and technological proposals have been developed to reduce the negative impact on the environment.

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Аңдатпа. Мақала су ресурстарының барлық жерде ластануы байқалатын «ТГПК» ЖШС базасында Текелі тау-кен өңдеу кешенінің әрекет ету ауданында ҚР Жетісу облысының Текелі өнеркәсіптік қаласының өзекті мәселесіне арналған. Оның су бассейнінің негізгі ластаушысы тау-кен өңдеу кешенінің алаңдарында шоғырланған жұмыс істеп тұрған кәсіпорынның су қалдықтары болып табылады. Қаратал өзенінің суы өзен торабының ең тығыз аймағында орналасқан өнеркәсіптік тау-кен өндіруші кәсіпорындардың улы компоненттерімен ластануда. Бұл бірлескен орналасу газ тәрізді, сұйық және қатты қалдықты заттардың өзен ағысына үздіксіз енуіне ықпал етеді. Осының нәтижесінде Жетісу өңірінің жер үсті сулары сапасының нашарлау сипаты орнықты үрдіске айналуда. Су қалдықтарын тазартудың сорбциялық әдістерін дамыту үшін ең көп таралған және қолжетімді бентониттерді қолдану қызығушылық тудырады. Алайда, белсендірусіз табиғи күйде олар көбінесе сорбциялық қабілеті төмен, бұл олардың тұтынуының төмендеуіне әкеледі. Алюмосиликаттардың табиғи минералды шикізатынан сорбциялық сыйымдылығы жоғары белсендірілген сорбенттерді алу қажеттілігі туындады. Бұл жұмыстың мақсаты – табиғи сорбент-бентонитті қолдана отырып, ауыр металдарды өнеркәсіпте тазартудың инновациялық технологиясын жасау. Әдістер: статистикалық әдістер, судың ластану индексін есептеу әдістемесі бойынша жер үсті суларының химиялық көрсеткіштерін бағалау. Аймақта жер асты және жер үсті сулары ауыр металдармен экологиялық ластануда. Бұған пайдалану әдістерінің жетілмегендігі – полиметалл кендерінің кен орындарын өндіру және өңдеу, сондай-ақ тау жыныстарының үздіксіз жиналатын үйінділері мен байыту қалдықтары тікелей өндірістік аумаққа әкеледі. Оларға химиялық элементтер мен олардың қосылыстарымен ұсынылған ағынды сулар мен дренажды сулардың әсеріне байланысты болады. Нәтижелер: өнеркәсіп орындарын ауыр металдардан (Zn^{2+}, Pb^{2+}, Cu^{2+}) қышқыл-сілтілі активтендірілген бентониттермен тазартуға тәжірибелік-өнеркәсіптік сынақтар қоршаған ортаға кері әсерін азайтуда экологиялық және технологиялық ұсыныстарды анықтады.

Негізгі сөздер: *алюмосиликатты-бентониттер, сорбциялық сыйымдылық, кәсіпорынның промстоктары, ауыр металдардың сорбциясы, улытты заттар, Қаратал өзені.*

Эффективность очистки промстоков природными сорбентами-бентонитами

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Аннотация. Статья посвящена актуальной проблеме промышленного города Текели Жетысукской области РК в районе действия Текелийского горно-перерабатывающего комплекса, на базе ТОО «ТГПК», где наблюдается повсеместное загрязнение водных ресурсов. Основным загрязнителем его водного бассейна являются промстоки действующего предприятия, сконцентрированного на площадках горно-перерабатывающего комплекса. Вода реки Каратал испытывает интенсивную нагрузку от токсичных компонентов промышленных предприятий горнодобывающей отрасли, которая расположена в зоне наиболее густой речной сети. Такое совместное расположение способствует к тому, что вещества с газообразными, жидкими и твердыми отходами неизбежно попадают в речную сеть. В результате этого характер ухудшения качества поверхностных вод Жетысуского региона становится устойчивой тенденцией. Существенный интерес для развития сорбционных методов очистки промстоков представляет применение алюмосиликатов – бентонитов, как наиболее распространенных и дешевых. Однако в естественном состоянии без активации они зачастую не обладают высокой сорбционной способностью, что влечет повышенный их расход. Возникла необходимость получения активированных сорбентов с более высокой сорбционной емкостью из природного минерального сырья алюмосиликатов. Целью данной работы является разработка иновационной технологии очистки промстоков от тяжелых металлов с использованием природного сорбента – бентонита. Методы: Статистические методы, оценка по химическим показателям поверхностных вод по методике, для расчета индекса загрязненности воды. В регионе подземные и поверхностные воды находятся в состоянии экологического загрязнения тяжелыми металлами. К этому приводят как несовершенство способов эксплуатации – добыча и переработка месторождений полиметаллических руд, так и непрерывно накапливаемые отвалы горных пород и хвосты обогащения непосредственно на производственных площадках. На них накладывает отпечаток влияние сточных и дренажных вод, представленных химическими элементами и их соединениями. Результаты: опытно-промышленные испытания по очистке промстоков от тяжелых металлов (Zn^{2+}, Pb^{2+}, Cu^{2+}) кислотно-щелочноактивированными бентонитами определили эколого-технологические предложения по снижению негативного воздействия на окружающую среду.

Ключевые слова: *алюмосиликаты-бентониты, сорбционная емкость, промстоки предприятия, сорбция тяжелых металлов, токсиканты, река Каратал.*

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