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ХИМИЧЕСКИЕ ПРЕВРАЩЕНИЯ ПЫЛИ РУКАВНЫХ ФИЛЬТРОВ ЗАВОДА ЖБИ В ВОДНЫХ РАСТВОРАХ

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Соединения никеля и меди являются одними из самых опасных загрязнителей окружающей среды, и их удаление из сточных вод является важной задачей. Для их извлечения из водных сред применяют традиционные методы, такие как ионный обмен, реагентная очистка, мембранная фильтрация, адсорбция. В последнее время для очистки водных сред от ионов тяжелых металлов все чаще предлагается использовать отходы различных производств, например, шлаки, шламы, золу, пыль и т.д. В работе рассматривалась возможность очистки сточных вод от ионов Cu^{2+} и Ni^{2+} с использованием в качестве реагентов пыли из рукавных фильтров производства железобетонных изделий. Определено, что пыль рукавных фильтров имеет тонкодисперсную структуру и содержит значительное количество силикатов различного состава. В работе описаны химические превращения, протекающие в водной среде при добавлении к ней пыли рукавных фильтров завода ЖБИ. Приведены некоторые реагенты, коагулянты, сорбционные материалы, которые могут быть использованы для очистки сточных вод от ионов тяжелых металлов. Указаны значения произведений растворимости образующихся соединений – гидроксида меди, гидроксида никеля, карбоната гидроксомеди. Приведены результаты гранулометрического, рентгенофазоваго и химического анализов пыли рукавных фильтров. Согласно проведенным исследованиям, эффективность очистки модельного раствора от ионов тяжелых металлов при этом составляет не менее 99,3%.

Ключевые слова: пыль рукавных фильтров, водная среда, химические превращения, эффективность очистки

CHEMICAL TRANSFORMATIONS OF DUST OF BAG FILTERS OF THE REINFORCED CONCRETE PLANT IN AQUEOUS SOLUTIONS

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Nickel and copper compounds are among the most dangerous environmental pollutants and their removal from wastewater is an important task. To extract them from aqueous media, traditional methods are used, such as ion exchange, reagent purification, membrane filtration, and adsorption. Recently, for the purification of aqueous media from heavy metal ions, it is increasingly proposed to use waste from various industries, for example, slag, sludge, ash, dust, etc. The paper considered the possibility of wastewater treatment from Cu^{2+} and Ni^{2+} ions using dust from bag filters in the production of reinforced concrete products as reagents. It was determined that bag filter dust has a fine structure and contains a significant amount of silicates of various compositions. The paper describes the chemical transformations that occur in the aquatic media when dust from the bag filters of the reinforced concrete plant is added to it. Some reagents, coagulants, sorption materials that can be used to purify wastewater from heavy metal ions are given. The values of the solubility products of the formed compounds - copper hydroxide, nickel hydroxide, hydroxomeric carbonate are indicated. The results of granulometric, X-ray phase and chemical analyzes of bag filter dust are presented. According to the studies, the efficiency of purification of the model solution from heavy metal ions in this case is at least 99.3%.

Key words: bag filter dust, aqueous environment, chemical transformations, purification efficiency

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INTRODUCTION

Heavy metal compounds contained in the wastewater of many industries are dangerous for environmental objects. Due to their high toxicity, carcinogenicity, mutagenicity, polluting effects on terrestrial ecosystems, in addition to the negative impact on animals, birds, fish fauna, in the end, it also poses a huge risk to human health. In recent years, awareness of the danger of pollution of water bodies and its far-reaching consequences has been growing all over the world.

Among the various heavy metals, nickel and copper are among the most toxic ones. Their danger can be judged by the values of maximum permissible concentrations (MPC) and the limiting sign of harmfulness (LSH) (Table 1).

Salts of nickel (II) slow down the action of oxidative enzymes, change the chemical properties of RNA and nucleoproteins during complex formation, and have a carcinogenic effect. Nickel causes cancer of the nasopharynx, bronchi, dermatitis, impaired hematopoiesis, embryotoxicosis, exhibits mutagenic and teratogenic effects.

Copper compounds cause liver and kidney dysfunction, nervous system disorders, muscle pain, and mutagenic effects. Poisoning with copper compounds is often combined.

Therefore, wastewater purification from heavy metal ions and preventing them from entering the environment is an urgent task. In recent years, waste from various industries and various etiologies has been increasingly used for wastewater treatment [1-9]. At the same time, many wastes successfully compete with the chemicals used in the practice of water purification [10-16].

Table 1

Characteristics of harmful substances

***Таблица 1.* Характеристика вредных веществ**

Metal	Water objects of economic and drinking and cultural and household water use		Water objects of fishery importance	
	MPC, мg/dm^3	LSH	MPC, mg/dm^3	LPH
Copper	1.0	organolept.	0.001	tox.
Nickel	0.02	sanit.-tox.	0.01	tox.

At Fig. 1, 2 as an example, the methods and reagents used in the practice of water purification from heavy metal ions are indicated.

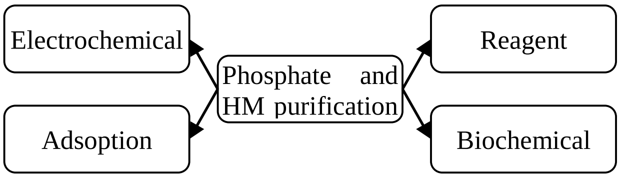


Fig. 1. Purification of phosphate and metal-containing wastewater
Рис. 1. Очистка фосфат- и металлсодержащих сточных вод

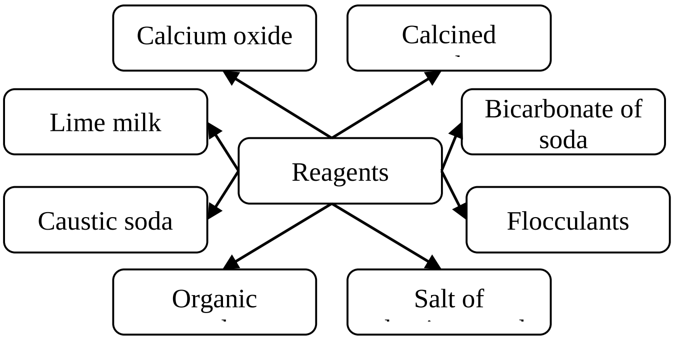


Fig. 2. Chemical substances used in water treatment
Рис. 2. Химические вещества, используемые в водоочистке

Purification from Ni^{2+} and Cu^{2+} ions proceeds due to the formation of poorly soluble compounds, the solubility product of which (SP) is indicated in Table 2 [1].

Table 2

Solubility products of precipitation
***Таблица 2*. Произведения растворимости осадков**

Substance	$Cu(OH)_2$	$Ni(OH)_2$	$(CuOH)_2CO_3$
SP	$2.2\cdot10^{-20}$	$8.7\cdot10^{-19}$	$1.70\cdot10^{-34}$

The limiting factor in the use of traditional reagents is the high cost of the latter, the need for constant renewal, the use of reagent facilities for the preparation of solutions and suspensions of the appropriate concentration of the active reagent, etc., which leads to an increase in the cost of the purification process.

In recent years, wastewater from different industries of various etiology, as well as natural mineral compounds, has been increasingly used for wastewater purification [2-10]. At the same time, many wastes successfully compete with chemical reagents used in the practice of water purification [11-17].

Sorption methods are often used for deep purification. At Fig. 3 substances frequently used in sorption purification are given [18-40].

Industrial wastes are of particular interest. They are generated in large volumes and their disposal at industrial waste landfills leads to the rejection of large areas of land. One of the ways to use industrial production wastes is their use as reagents for the removal of various pollutants from wastewater. In particular, we have previously shown the possibility of using waste from the construction industry to extract heavy metal ions, dyes, and other pollutants. It has been shown; in particular, that bag filter dust effectively extracts heavy metal ions from model wastewater [11-16]. To assess the further possibility of using bag filter dust in water purification, we considered its supposed transformations in the aquatic media.

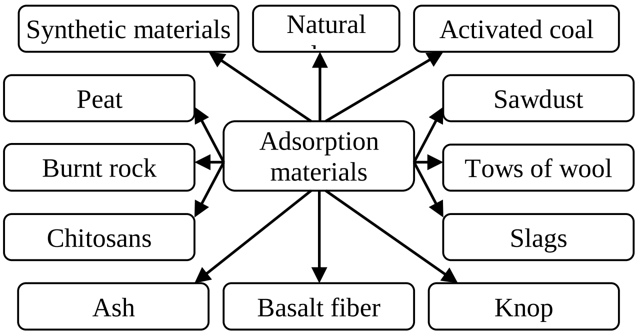


Fig. 3. Alternative reagents used in sorption wastewater purification
Рис. 3. Альтернативные реагенты, используемые в сорбционной очистке сточных вод

MATERIALS AND METHODS

Dust from bag filters of the ZhBI-4 plant (Belgorod) was used in the work. The size of the dust particles, determined using a standard set of sieves, did not exceed 1.4 mm. For research, a fraction with sizes from 0.315 to 1.4 mm was selected.

The relief structure of the particle surface was studied using electron microscopy with a Hitachi-8-800 instrument combined with a personal computer and a high-resolution scanning electron microscope of the TESCAN MIRA 3LMU brand.

The phase composition of the dust was determined using an ARL 9900 Intellipower Workstation instrument using a tube with a Co-anode.

Model solutions containing Ni^{2+} and Cu^{2+} ions were prepared by dissolving reagents $CuSO_4\cdot5H_2O$ (chemically pure) and $NiSO_4\cdot7H_2O$ (chemically pure) in distilled water.

Purification of model solutions was carried out by adding a certain mass of dust to 100 cm^3 of the solution placed in a conical flask with a capacity of 250 cm^3, after which stirring was carried out, followed by filtration through a paper filter. The content of Ni^{2+} and Cu^{2+} ions in the filtrate was determined by the photocolorimetric method. The purification efficiency was calculated using the standard formula:

$$E = (Cin - Cfin)/Cin\cdot100,$$

E – purification efficiency, %; Cin and Cfin – initial and final concentrations of pollutants in solution, mg/dm^3.

RESULTS AND ITS DISCUSSION

In accordance with X-ray phase analysis, anhydrite, hartrurite, larnite, albite, gypsum, brownmillerite, grossite, belite, aliite, tricalcium aluminoferrite, tetracalcium silicate, calcite, and quartz were found in the dust of bag filter composition.

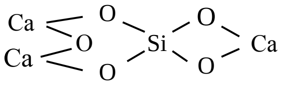
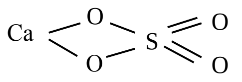
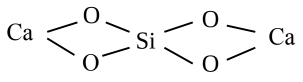
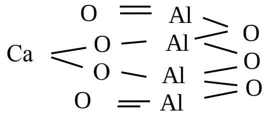
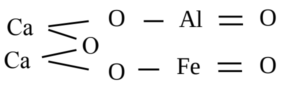
The mineral composition of dust of bag filter is represented by the following compounds: C_3S – alite: $3CaO \cdot SiO_2$ (d, A^o = 3.043; 2.978; 2.898; 2.64; 2.191; 1.771); C_2S – belite: $2CaO \cdot SiO_2$ (d, A^o = 2.898; 2.784; 2.614; 2.452; 2.404; 2.191); C_3A – tricalcium aluminate: $3CaO \cdot Al_2O_3$ (d, A^o = 2.702; 1.937); C_4AF – tetracalcium aluminoferrite: $4CaO \cdot Al_2O_3 \cdot Fe_2O_3$ (d, A^o = 7.661; 3.678; 2.784; 2.67; 2.056); SiO_2 – quartz (d, A^o = 4.181; 3.351; 2.452; 2.327; 2.28; 1.983; 1.826); $CaCO_3$ – calcite (d, A^o = 3.043; 2.503; 1.937).

Graphic formulas of some substances are presented in Table 3.

Table 3

Characterization of some compounds

***Таблица 3*. Характеристика некоторых соединений**

№	Name	Formula	Graphic formula
1	Hartrurite	Ca_3SiO_5	
2	Anhydrite	$CaSO_4$	
3	Larnite	Ca_2SiO_4	
4	Grossite	$CaAl_4O_7$	
5	Brownmillerite	$Ca_2(Al,Fe^{3+})_2O_5$	

Such compounds as $3CaO \cdot SiO_2$ – tricalcium silicate, $CaO \cdot SiO_2$ – calcium metasilicate, $2CaO \cdot SiO_2$ – calcium orthosilicate are also found in the selected dust samples.

When DBF enters the aqueous environment, the following processes may occur:

$$Ca_2SiO_4 \rightarrow 2Ca^{2+} + SiO_4^{4-}$$

$$3CaO \cdot SiO_2 + 5H_2O \rightarrow$$

$$\rightarrow 3CaO \cdot 2SiO_2 \cdot H_2O + 3Ca(OH)_2 + Q$$

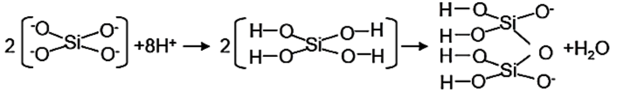
$$3CaO \cdot SiO_2 + 3H_2O \rightarrow$$

$$\rightarrow 2CaO \cdot SiO_2 \cdot 2H_2O + Ca(OH)_2 + Q$$

$$2(2CaO \cdot SiO_2) + 3H_2O \rightarrow$$

$$\rightarrow 3{,}3CaO \cdot 2SiO_2 \cdot 2{,}3H_2O + 0{,}7Ca(OH)_2$$

During the dissociation of compounds such as Ca_3SiO_5; Ca_2SiO_4 Ca_3SiO_5 $CaAl_4O_7$; $Ca_2(Al, Fe^{3+})_2O_5$ and others, solution acquires an alkaline reaction due to the transition of Ca^{2+}, Na^+ ions into the solution. When dissolving silicates of various composition (SiO_4^{4-}); $(Si_3O_8)^{4-}$ due to the ability of silicate ions to polycondensate, the formation of network structures with a large surface having a negative charge is possible:



The formation of structures with a large negative surface in the solution promotes the process of approaching the positively charged metal ions Ni^{2+}, $NiOH^+$, $[Ni(H_2O)_2]^{2+}$ and negative surface charges, which leads to faster removal of heavy metal ions from the solution.

Thus, when dust from bag filters is added to the aqueous medium, the purification from heavy metal ions can proceed according to the reagent mechanism due to the formation of poorly soluble hydroxides such as $Ni(OH)_2$, $Cu(OH)_2$ and due to the probable sorption interaction, since due to the high surface imperfection dust particles have sorption activity (Fig. 4).

To confirm the possibility of using dust from bag filters of reinforced concrete products plants in water purification, we purified model solutions containing Ni^{2+} and Cu^{2+} ions at concentrations of 10 mg/dm^3.

The studies were carried out under static conditions at a temperature of t = 20 °C on model solutions containing metal ions, stirring time τ = 20 min. After stirring, the suspension was quantitatively transferred to a settling cylinder p and subjected to sedimentation for 30 min. Then, a layer of the clarified liquid was taken with a pipette and analyzed for the residual content of Ni^{2+} and Cu^{2+} ions.

It has been established that the purification efficiency for both studied substances sharply increases when even small amounts of dust are added to the solution. This is due to the fact that with the introduction of dust, the pH of the solution greatly increases and favorable conditions are created for the formation of poorly soluble hydroxides.

Thus, when only 0.2 g of dust is added per 1 liter of solution, the purification efficiency reaches 73% for Ni^{2+} and 68% for Cu^{2+}. Then the increase in efficiency slows down and with the addition of 0.5 g, the efficiency increases by only 15%. It is most likely due to the fact that in the first time since the formation of $Ni(OH)_2$ and $Cu(OH)_2$ precipitates, the concentrations of hydroxide ions and metal ions in the solution drop sharply, the product of their concentrations decreases, and in order to achieve the solubility product of hydroxides in the same solution, it is necessary to add new portions of dust. With the addition of new portions of dust, the purification efficiency increases, but its growth is less and less intense, which can be explained by a decrease in the concentrations of metal ions in solutions.

Due to the fact that substances contained in the solid phase (dust) and liquid (model solution) take part in the purification process, many physical and chemical processes proceed with the participation of the phase interface. For such processes, the magnitude of the phase separation surface or contact surface is of great importance. In our case, such a contact surface is the surface of dust particles. With the participation of this surface, the processes of dissolution and crystallization, adsorption and desorption proceed. Since the surface of the solid phase in the case of non-porous and low-porous sorbents depends to a greater extent on the particle size, it was of interest to study the dependence of the purification efficiency on the fineness of the applied dust.

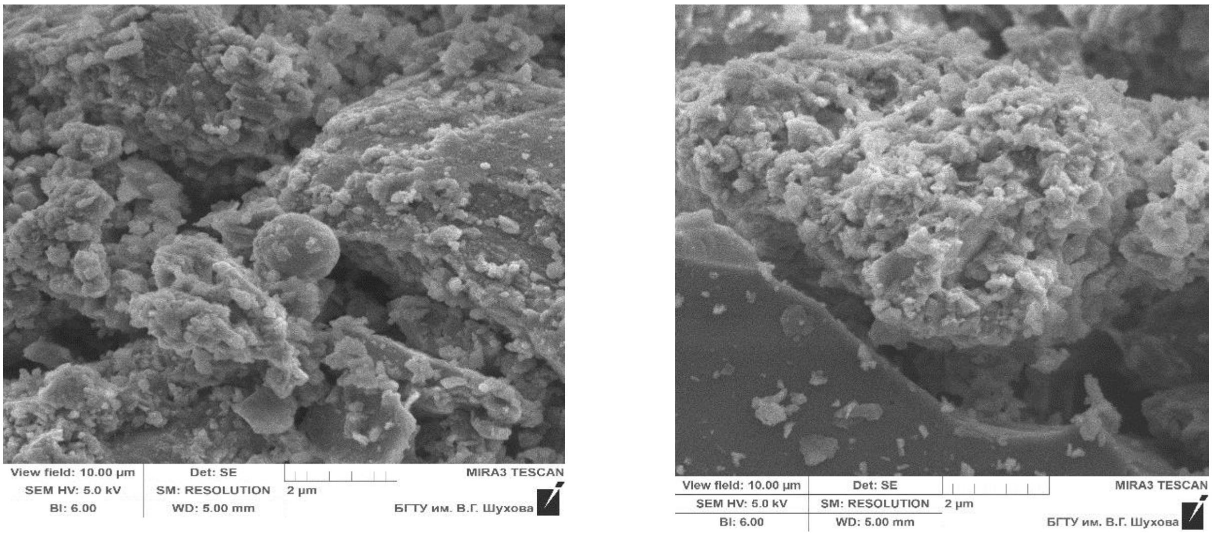


Fig. 4. Fragments of DBF sections
Рис. 4. Фрагменты участков ПРФ

Dependences of the efficiency of the purification process of model solutions on the used sorbent fraction were obtained, i.e. E(%) = f(d). In this case, model solutions were used, where $[Ni^{2+}]$init = $[Cu^{2+}]$init – 10 mg/dm^3; dust consumption was 1 g/dm^3; duration of the purification process - 20 min.

According to the data obtained, the largest amount of metal ions is removed from the solution by a dust fraction of less than 0.1 mm, which is explained by the smallest size of the average diameter and, as a result, a larger specific surface area.

Thus, the possibility of using industrial waste such as bag filter dust as a reagent for removing heavy metal ions from aqueous media was considered. The dust composition was determined by X-ray phase analysis. It was revealed that in its composition there are inorganic compounds of calcium, silicon and aluminum, which, when dissociated in water, contribute to the formation of the corresponding hydroxides of heavy metals that precipitate. Another alternative mechanism for the removal of heavy metal ions is adsorption on the surface of the sorption material.

The authors declare the absence a conflict of interest warranting disclosure in this article.

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