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ИЗВЕСТИЯ

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РЕСПУБЛИКИ КАЗАХСТАН
АО «Институт топлива, катализа и
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MODIFIED SORBENTS AND THEIR APPLICATION FOR EXTRACTION OF METAL IONS

Abstract. The data on accumulated mining waste in the territory of Kazakhstan are presented. Their processing has become very important in our time. The structure of surface-modified carbon materials with a grafted layer is given. New modified carbon sorbents were obtained, oleum was used as a modifying agent. It was revealed that the capacitive characteristics of the modified sorbent are significantly higher than that of the original one, which will make it possible to more efficiently extract the metal ion from wastewater and technogenic formations. Sorption is an effective method of extracting valuable components from wastewater and man-made formations. This method allows you to recover valuable substances with a high degree of wastewater treatment, which can be purified to the maximum permissible concentration of pollutants and then used in technological processes or in recycling water supply systems. Processing involves ores characterized by a low content of valuable components, fine dissemination and similar technological properties of minerals. The existing technologies for the development of minerals make it possible to use only a small part of the valuable mineral mass extracted from the subsoil, and the rest forms waste, which, as it accumulates and is stored, becomes one of the most significant factors of anthropogenic changes in the environment.

Key words: coal, sorbents, modification, sorption isotherm, sorbent capacity, oleum.

Introduction. Kazakhstan has the richest raw material base for the production of sorption materials, which makes it possible to obtain sorbents for various purposes with an optimal combination of price and quality. Recycling of mining waste has become a key factor in ensuring the country's environmental safety. Many of them contain toxic substances of the first and second hazard classes, which are priority pollutants, have a negative effect on nearby territories, changing the chemical composition of soils, disrupting the unity of the geochemical environment and living organisms, and on the entire coastal-marine zone due to the transboundary transfer of pollutants by waters and air currents.

Wood (in the form of sawdust), charcoal, peat, peat coke, bituminous and brown coals can be used as raw materials for obtaining carbon sorbents (CS). To obtain US with high strength properties and a large volume of fine pores, such unconventional materials are used as shells of various types of nuts, fruit pits, etc. [1,2].

In the sorption extraction of a metal ion, ion exchange processes are of great importance [3]. The developed porous structure and the presence of basic and acidic functional groups on the surface of sorbents are necessary conditions for the extraction of metal ions from industrial solutions. Functional groups (carboxyl, phenolic, lactone, etc.) are capable of exchanging protons or hydroxyl groups for metal ions or their complexes in solutions.

The authors have obtained US from bituminous and brown coals with good capacitive characteristics [4]. To increase the sorption capacity of US in relation to the extracted components (metals, mercury vapor, fluorine, ammonia, biological objects, etc.), modification of carbon sorbents is used [5]. This

problem is solved by purposefully affecting the surface properties of the US for the formation of various sorption centers, changing the bulk properties of materials and chemical properties of the surface, primarily its hydrophilicity or hydrophobicity [6]. This modification of carbon sorbents makes it possible to obtain a relatively new class of carbon sorbents, which are a solid, on the surface of which an extremely thin, usually molecular, layer of chemical compounds is fixed. In this case, the "substance on a carrier" system is often a new material with a number of properties that neither the carrier nor the sorbed compound possessed. The fixation of the active chemical component on the surface of the carrier can be carried out due to physical or chemical interactions, the latter is preferable, since it makes it possible to achieve a significantly greater resistance of the resulting materials to various influences and stability of action. The chemical properties of such surface-modified carbon sorbents are determined by the nature of the fixed compound, while the physical and mechanical properties of the carrier. In the structure of surface-modified carbon materials, it is always possible to distinguish common constituent elements: the anchor group is responsible for fixing the grafted compound; leg - is a group that separates the grafted compound from the surface; function - a group (groups) in which the properties of the grafted compound (adsorption) are concentrated.

The grafted layer can be monomeric or polymeric, dense or loose, mono - and polyfunctional. The distribution of the grafted molecules can also be different: statically random, insular, or regular. The grafted layer can be a set of several types of functional groups. To obtain chemically uniform grafted layers, it is necessary to choose the correct reagents and modification conditions. There are many ways to modify carbon materials. The most common oxidative treatment can be carried out in both liquid and gaseous environments. For this purpose, high-temperature oxidation of the carbon material surface with gaseous oxidants (CO_2, water vapor, atmospheric oxygen), ozone oxidation [7], oxidation in oxygen and isobutylene plasma [8], treatment with mineral acids or their mixtures, hydrogen peroxide, dichromate or permanganate are used. in concentrated aqueous solutions, etc. [9,10]. Oxidants change the spatial structure and molecular structure of coal [11-12]. Surface oxidation occurs with the formation of various functional groups and the incorporation of anions into the interplanar spaces of the crystal structure of coal. During oxidation with liquid oxidizing agents, for example, nitric acid, a chemical interaction of surface carbon atoms with an oxidizing agent occurs, as a result of which oxygen-containing functional groups are formed. Oxidants change the spatial and molecular structure of coal, i.e. convert it to structurally modified coal.

Oxidative modification increases the specific surface area (from 2 to 30 times) and the total pore volume (up to 1.9 cm^3 / g), and also makes it possible to vary the proportion of micropores and the pore size distribution, i.e. to obtain predominantly micro- or mesoporous CS.

Among the products of chemical modification of coal, sulphonated coal (sulfocoal) is of great importance, which has been used for many years as a cation exchanger for desalting water in the energy industry and for other purposes [13-14]. Sulfonation improves the mechanical and cation exchange properties of coal.

The use of cation exchangers in various technological processes leads to a radical simplification and improvement of production schemes, which determines the effectiveness of their use in the chemical, mining and processing industries. Due to the simplicity of its preparation, sulfocarbon successfully competes with ion-exchange resins, but is inferior to them in terms of chemical resistance.

Sulfonated carbon is a polyfunctional cation exchanger, in the structure of which, in addition to the strongly acidic sulfo group, there are also weakly acidic carboxyl groups and phenolic hydroxyl.

Dissociation of phenolic hydroxyl groups depends on the pH of the medium and becomes noticeable at pH = 9, carboxyl groups at pH greater than 5, and sulfo groups completely dissociate at all pH values.

The exchange capacity of sulfo coal for certain stages of metamorphism is directly proportional to the content of vitrinite group microcomponents in the initial coal. A significant release of volatile substances on a hot mass indicates the presence of a developed periphery in their structure, as well as free or useful hydrogen. It is known that with an increase in the development of the periphery, the reactivity of coal increases. The lower the density and the greater the porosity of the original coal, the better the sulfonation process occurs. That is why the authors propose to sulfonate coal obtained from brown coals of the Shubarkul basin.

The availability and low cost of brown coal attracts the attention of researchers. The choice of raw materials was determined by its natural and physicochemical properties: initial porous structure, high yield of volatile substances (40%), low ash and sulfur content (0.5%). Thermally exposed brown coals have a developed porous structure, which contains pores of all sizes - from micropores to visible large pores. The presence of a branched system of transport pores provides good surface accessibility to the molecules of the modifying agent.

The carbon sorbent obtained by thermal treatment according to the classical scheme, including carbonization and steam-gas activation, was subjected to modification.

As a modifying agent, oleum was used with an SO_3 concentration of 20%, at a ratio: CS: oleum = = (1: 1, 1: 2, 1: 3). Based on the results of previous studies, the optimal conditions for the sulfonation of CS were determined, under which the process was carried out. Before treating the CS with oleum, it was additionally pretreated with dichloroethane. The role of the dichloroethane solvent was to weaken interfragment and intrafragment non-bonded bonds (hydrogen and sorption bonds, van der Waals cohesion forces, etc.), loosening the structures, and opening pores

The treatment of the carbon sorbent with dichloroethane increases the diffusion rate of the reagent (oleum) deep into the coal substance, therefore, increases the reaction rate, and also affects the degree of elimination of peripheral fragments from the coal substance molecule.

Dichloroethane is an inert solvent with respect to the sulfonating agent. Dichloroethane solvates fragments of organic matter of coal, weakens intermolecular association, and releases reaction centers [15-17].

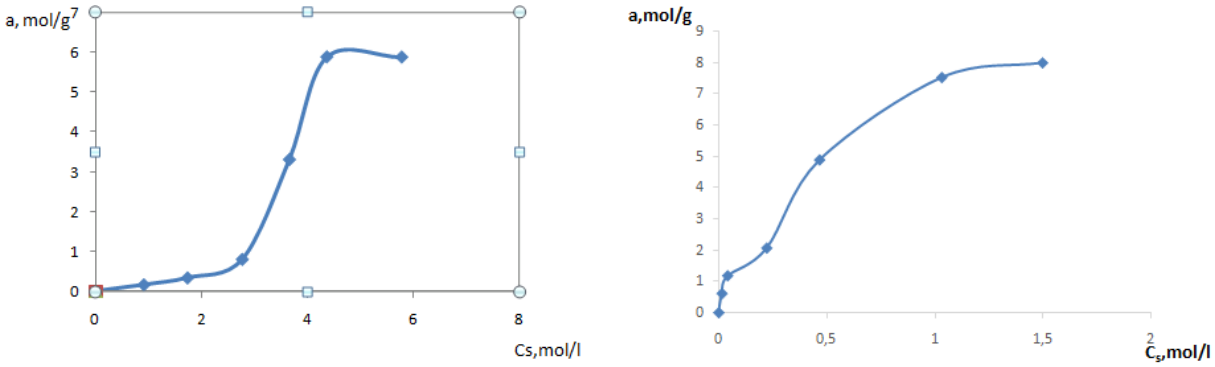
The physical and mechanical properties and parameters of the porous structure of the carbon sorbent (C) and modified (MC) are given in table.

Table 1 – Characterization of carbon sorbents

Brand sorbent	Bulk density, g / cm^3	Moisture contents, %	Mechanical strength,%	Total porosity by water, cm3 / g	Activity	
					by iodine,%	MG, mg / g
У	0,610	1,61	86	0,69	65,0	13,1
МУ	0,570	1,59	72,1	0,75	55,0	13,8

Under static conditions at a temperature of 120 °C, studies were carried out on the adsorption of the copper (II) ion on the studied sorbents C and MC at optimal pH values (2-14) in equilibrium conditions in the concentration range 5-20 mg / L The contact time of the sorbent with salt solutions was 2 hours.

Based on the results of studies of the adsorption of metal ions from aqueous solutions by natural and modified forms of sorbents, adsorption isotherms were constructed, which are one of the main criteria for assessing the adsorption properties of the studied sorbents and determine the dependence of the adsorbent activity on the adsorbate concentration under equilibrium conditions (figure).



а) в)

Isotherms of metal adsorption by the investigated samples: a - natural coal (C); в - modified coal (MC)

Table 2 – Capacity of sorbents for metal ions

Sorbent	Sorption capacity for ions, mg / g
	Copper (II)
C	5,88
MC	8,2

The values of sorption capacities for copper on the sorbent C and MC are given in table 2. Figure and table 2 that the capacity of the modified sorbent (MC) in relation to the metals under study is higher than that of the carbon sorbent (C).

During the treatment of the carbon sorbent with oleum, oxidation reactions occur, as a result of which carboxyl and phenolic groups are formed. The formation of acidic groups SO_3H, COOH, OH made it possible to obtain a sorbent with a high sorption capacity with respect to metal ions.

The studies carried out have shown that sorbents have a significant sorption capacity in relation to heavy metal ions, which makes it possible to use these MC in the technology of additional purification of wastewater and the extraction of valuable components from technogenic formations.

Conclusion**.** The authors believe that replacing C with MC will significantly reduce the consumption of the sorbent, therefore, improve the economic performance of the proposed technology. This technological scheme is distinguished by its simplicity of design, the use of traditional equipment and can be applied at the enterprises of the mining, chemical and petrochemical industries.

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МОДИФИЦИРЛЕНГЕН СОРБЕНТТЕР ЖӘНЕ ОЛАРДЫ МЕТАЛЛ ИОНДАРЫН АЛУҒА ҚОЛДАНУ

Аннотация. Қазақстан аумағындағы жинақталған тау-кен қалдықтарын өңдеу туралы мәліметтер келтірілген. Қазіргі кезде оларды өңдеу өте маңызды. Беттік қабаты бар өзгертілген көміртекті материалдардың құрылымы келтірілген. Жаңа модификацияланған көміртекті сорбенттер алынды, модификациялаушы зат ретінде олеум қолданылды. Модифицирленген сорбенттің сыйымдылық сипаттамалары бастапқыға қарағанда едәуір жоғары екендігі анықталды, бұл металл ионын ағынды судан және техногендік түзілімдерден тиімді шығаруға мүмкіндік береді. Сорбция – ағынды судан және өндірістік қабаттардан бағалы компоненттерді алудың тиімді әдісі. Бұл әдіс ағынды суды тазартудың жоғары дәрежелі құнды заттарды қалпына келтіруге мүмкіндік береді, оларды ластаушы заттардың рұқсат етілген шекті концентрациясына дейін тазартуға болады, содан кейін технологиялық үдерісте немесе сумен жабдықтау жүйелерін қайта өңдеу кезінде қолдануға болады. Өңдеуге бағалы компоненттердің аз мөлшері, ұсақ таралуы және минералдардың ұқсас технологиялық қасиеттерімен сипатталатын кен жатады. Пайдалы қазбаларды игерудің қолданыстағы технологиялары жер қойнауынан алынған құнды минералды массаның тек аз бөлігін ғана пайдалануға мүмкіндік береді, ал қалғаны қалдық түзеді, олар жинақтау және сақтау ретінде қоршаған ортадағы антропогендік өзгерістердің маңызды факторларының біріне айналады.

Түйін сөздер: көмір, сорбенттер, модифицирлеу, сорбциялық изотерма, сорбент сыйымдылығы, олеум.

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МОДИФИЦИРОВАННЫЕ СОРБЕНТЫ И ИХ ПРИМЕНЕНИЕ ДЛЯ ИЗВЛЕЧЕНИЯ ИОНОВ МЕТАЛЛОВ

Аннотация. Приведены данные по накопленным на территории Казахстана отходам горного производства. Их переработка приобрела в наше время важное значение. Дано строение поверхностно-модифицированных углеродных материалов с привитым слоем. Получены новые модифицированные углеродные сор-

бенты, в качестве модифицирующего агента использовался олеум. Выявлено, что емкостные характеристики модифицированного сорбента значительно выше, чем у исходного, что позволит с большей эффективностью извлекать ион металла из сточных вод и техногенных образований. Эффективным методом извлечения ценных компонентов из сточных вод и техногенных образований является сорбционный метод. Данный метод позволяет рекуперировать ценные вещества при высокой степени очистки сточной воды, которая может быть очищена до предельно допустимых концентраций загрязняющих веществ и затем использована в технологических процессах или в системах оборотного водоснабжения. В переработку вовлекаются руды, характеризующиеся низким содержанием ценных компонентов, тонкой вкрапленностью и близкими технологическими свойствами минералов. Существующие технологии освоения полезных ископаемых позволяют использовать лишь небольшую часть извлекаемой из недр ценной минеральной массы, а остальная часть образует отходы, которые по мере накопления и хранения становятся одним из наиболее значимых факторов антропогенных изменений окружающей среды.

Ключевые слова: уголь, сорбенты, модифицирование, изотерма сорбции, емкость сорбентов, олеум.

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