ANNOTATION

dissertation on the topic "Sorption purification of water from heavy metal ions using sulfocarbon and modified anionite" for the degree of Doctor of Philosophy (PhD) in the educational program 8D05311-"Chemistry"

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Research topic: "Sorption purification of water from heavy metal ions using sulfocarbon and modified anionite"

Research objective:

Development of new sorption materials based on sulfonated natural carbon and anion exchanger modified with citric acid to reduce the negative impact of heavy metal cations on water bodies.

Research objectives:

1. Synthesis and study of the physicochemical properties of the surface of sorbents based on sulfuric acid-modified coal from the Shubarkul deposit and anion exchanger modified with citric acid (AB-17-8: $C_6H_8O_7$);

2. Calculate the thermodynamic characteristics of adsorption and the values of the energies of specific interactions on the original and modified anion exchanger and sulfonated carbon.

3. Identification of patterns of adsorption of heavy metal cations, comparative assessment of polarity, sorption capacity and efficiency of sorbents in relation to common heavy metal cations Cu(II), Ni(II), Hg(II) under the influence of the main natural factors t, °C, pH and quantitative presence of cations.

4. To offer recommendations on the use of sorbents based on modified carbon and anion exchanger for environmental water purification and sorption concentration of heavy metal cations.

Research methods:

theoretical: - data on the kinetics of sorption of heavy metal cations on sulfonic carbon, measurements of the ζ-potential and suspension effect may indicate a negatively charged surface of sulfonic carbon in the working range of pH 5.5-7, which is proven by the adsorption of SCM and acid-base indicators.

-Under optimal conditions, using the data of sorption isotherms of SCM on sulfonic coal, the values of the maximum sorption of individual cations and the sum of metal cations and, accordingly, the equilibrium constants of the sorption-desorption process of cations were determined. At pH in the range of 5.6-6.8, for

which elemental, X-ray diffraction and IR spectroscopic analyses were carried out; the solubility constants KS were determined.

empirical: The following modern physicochemical research methods were used in the work: IR spectroscopy, adsorption porosimetry, scanning electron microscopy spectrophotometry, thermogravimetry.

The main provisions submitted for defense (scientific hypotheses and other conclusions constituting novelty are proven):

1. The ability of coal modified with concentrated sulfuric acid (sulfonic coal) to sorb heavy metals (Cu (II), Ni (II), Hg (II)) is 1.3-1.4 times higher than the original coal.

2. The ability of the anionite modified with citric acid (AB-17-8: $C_6H_8O_7$) to sorb heavy metal cations (Cu(II), Ni(II), Hg(II)) is 1.2-1.7 times greater.

3. As a result of modification of the anionite (AB-17-8: $C_6H_8O_7$) with acid, the number of weak acid-base centers in the pKa 4.1-5.5 region increases, the readsorption capacity increases, special electrostatic interactions appear, and the surface increases.

Justification for the novelty and significance of the obtained results and their compliance with scientific development directions or state programs:

1. Scientific novelty:

- To study the effect of modifier application conditions on pore distribution and the activity of the obtained sorbents, a series of new sorbents of modified anionites and carbons were synthesized. For the first time, the approach of applying an active composition to the surface of an inert structured primary carrier, providing the necessary textural characteristics, thermal and mechanical stability of the sorbent is widespread anion exchanger and coals of the Shebarkul deposit.

- It is shown that the use of sulfuric acid as a modifier of the unique porous structure of the Shebarkul coals affects the formation and distribution of Me (II) cations on the surface: two types of sorption of Me (II) cations are observed: 0.5-3.0 nm in size, localized mainly inside the pores of sulfocoal, and larger particles (4-8 nm) located on the outer surface of sulfocoal.

- For the first time, the dependence of the sorption of heavy metal cations on sulfocoal and AB-17-8: $C_6H_8O_7$ on pH was studied. As a result, sorption isotherms of SCM and individual metal cations were constructed, approximated by regression equations. These isotherms are of the Langmuir type, related to the monomolecular type of adsorption.

- It is suggested that the difference in the molar ratio of components in the adsorbate from the stoichiometric one can serve as evidence of the adsorption of

metal cations on various active centers of sulfonic coal, the presence of which is shown by the Hammett indicator method.

2. Main results of the study:

- Treatment of Shubarkol coal with sulfuric acid H_2SO_4 and anion exchanger 8: $C_6H_8O_7$ with citric acid made it possible to obtain sorbents with high sorption capacity with respect to heavy metal cations.

- Under optimal conditions, according to the data of SCM sorption isotherms on sulfonic coal, the values of the maximum sorption of individual cations and the sum of metal cations and, accordingly, the equilibrium constants of the sorption-desorption process of cations were determined. At pH in the range of 6-8, for which elemental, X-ray diffraction and IR spectroscopic analysis; solubility constants KS were determined. It was suggested that the difference in the molar ratio of components in the adsorbate from the stoichiometric one may serve as evidence of adsorption of metal cations on various active centers of sulfocoal, the presence of which was shown by the Hammett indicator method.

- The dependence of the sorption of heavy metal cations on AB-17-8: $C_6H_8O_7$ and sulfocoal on pH was studied. As a result, sorption isotherms of SCM and individual metal cations were constructed, approximated by regression equations. These isotherms are of the Langmuir type, related to a monomolecular adsorption layer. Using the linearization of the initial section of the isotherm, the maximum sorption values of individual cations (a_∞ = 1.07 mmol/g) and the sum of metal cations (a_∞ = 1.14 mmol/g), as well as their sorption-desorption constants (K_{ind} = 388 and K_{sum} = 109.6) were determined. The maximum sorption value of the initial anion exchanger is 0.012 mmol/g, which is less than that of AB-17-8: $C_6H_8O_7$, which is an effective adsorbent for SCM.

- The possibility of adsorption extraction of SCM at a concentration of less than 0.002 mol/l and complete extraction (~100%) from water on AB-17-8: $C_6H_8O_7$ and sulfocarbon is shown.

3. Practical significance:

- When Shubarkol coal is treated with sulfuric acid H_2SO_4, the Me(II) sorption reaction occurs, resulting in the formation of phenolic carbon groups and acid groups $-SO_3H$, –COOH, –OH, which made it possible to obtain a sorbent with a high sorption capacity for heavy metal cations.

- The possibility of adsorption extraction of SCM at a concentration of less than 0.002 mol/l and complete extraction (~100%) from industrial waters on sulfocoal and AB-17-8: $C_6H_8O_7$ is shown.

- Thermodynamic characteristics of adsorption of heavy metal cations on sulfocarbon and AB-17-8:C6H8O7 were calculated

The results obtained will serve as a basis for creating a sorption system for the adsorption of heavy metals from water of natural and industrial origin, including for the purification of waste water from chemical, pharmaceutical and other enterprises.

4. The dissertation work corresponds to the priority direction of scientific development: "Rational use of natural resources, including water, geology, extraction and processing of minerals and hydrocarbons, new materials and technologies, safe products and designs."

The connection of the dissertation topic with scientific research work and various state programs. The work was carried out on the basis of state grant funding under the project BR24992867 "Development of resource-saving technologies for the development and management of water management and processing industries of Kazakhstan, creation of an innovative engineering center."

Description of the doctoral student's contribution to the preparation of each publication (the share of the dissertation author is indicated as a percentage of the total text):

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1. Control for selective sorption of heavy metals cations with anion exchanger AB-17-8 by modifying the surface with citrate groups. Mendeleev Communications. 34(5) 2024, P. 755-757. (co-authors Bektenov N.A., Serebryakov K.V., Elkin U.S., Gavrilenko M.A., doctoral student's share 80%)

5 articles published in journals recommended by the Committee for Quality Assurance in Science and Higher Education of the Ministry of Higher Education of the Republic of Kazakhstan:

1. Production of sulfocationite by modification of natural coal with concentrated sulfuric acid. News of the Academy of Sciences of the Republic of Kazakhstan. Series of Chemistry and Technology. No. 3. 2020.- P. 104-109. (co-authors Bektenov N.A., Gavrilenko M.A., Nurlybaeva A.N., doctoral student's share 80%)

2. Sorption of rare earth metals: literature review. Kazakhstan Chemical Journals No. 3. 2020.-P. 214-235. (co-authors: Bektenov N.A., 20%, Gavrilenko M.A. doctoral student's share 80%)

3. Modified sorbents and their application for extraction of metal ions. News of the National Academy of Sciences of the Republic of Kazakhstan. Series of Chemistry and Technology. №1. 2021.- P. 75-79. (co-authors Bektenov N.A., Gavrilenko M.A., doctoral student's share 80%)

4. Investigation of the sorption of heavy metals on modified coal. News of the National Academy of Sciences of the Republic of Kazakhstan. Series Chemistry and Technology. №4. 2022. – P. 118-125. (co-authors: Gavrilenko M.A., Bektenov N.A., Kudaibergenova R.M., Seitbekova G.A., doctoral student's share 80%)

5. Sorption purification of water from heavy metal ions using sulfougl. Chemical Journal of Kazakhstan №1. (81) 2023.- P. 75-82. (co-authors Gavrilenko M.A., Bektenov N.A., Kudaibergenova R.M., Baibazarova E.A., doctoral student's share 80%)

11 articles in the materials of international scientific and practical conferences, including those in the near and far abroad:

1. Pollution of water resources with various substances. "Digital Kazakhstan: digital lands rudyn zhahandyk trendteri zhane khalykaralyk tazhiribe." Halykaralyk gylymi praktikalyk conference matrialdary. 2019. B. 208-213. (co-authors Kambarova E.A., doctoral student's share 70%)

2. Effective methods for removing heavy metals with ion exchangers and modification of ion exchangers. Bulletin of TarSU named after M.Kh.Dulati. No. 27. 2019 P.24-32. (co-authors Bektenov N.A., Kambarova E.A., doctoral student's share 70%)

3. New chelating sorbents. IV All-Russian Scientific Symposium "Actual Problems of Theory and Practice of Heterogeneous Catalysts and Adsorbents" July 1-3, 2019. International Conference, Ivanovo-Suzdal. P.29-31 (co-authors Bektenov N.A., Kambarova E.A., doctoral student's share 50%)

4. Modification of anion exchanger with citric acid for efficient sorption International Symposium "Problems of Geology and Subsoil Development" Volume II, Russia-2020. P.357-358. (co-authors Bektenov N.A., Kambarova E.A., Gavrilenko M.A., doctoral student's share 60%)

5. Study of the possibility of wastewater purification from metal ions on new sorbents. International Symposium "Problems of Geology and Subsoil Development", Volume II, Russia-2020. P.348-349. (co-authors Bektenov N.A., Kambarova E.A., Gavrilenko M.A., doctoral student's share 40%)

6. Obtaining new promising complexing ion exchangers and their quantum-chemical calculations. Proceedings of the International scientific and practical conference "Actual problems of ecology and human life safety in the 21st century". Volume I. Taraz-2021, pp. 42-47. (co-authors Bektenov N.A., Sadykov K., Kasymbekova D., doctoral student's share 70%)

7. Creation of new polyfunctional ion exchangers and their application. "Fine Organic Synthesis-2021." Proceedings of the scientific conference, Almaty-

2021. pp. 19-21. (co-authors Bektenov N.A., Kambarova E.A., doctoral student's share 50%)

8. Prospects for the application of ion-exchange materials in the field of colorimetric methods of analysis. International scientific and practical conference. "Trends, prospects and innovative approaches to the development of chemical science, production and education in the context of globalization", dedicated to the 80th anniversary of the birth of the outstanding Kazakhstani scientist-internationalist - Academician of the NAS RK Ergozhin Edil Ergozhayevich. Almaty-2021zh. 87-90b. (co-authors Gavrilenko M.A., Bektenov N.A., Kambarova E.A., doctoral student's share 50%)

9. Sorption suction of lanthanum ion with new modified sorbents. Mechanics and Technology / Scientific journal 2023, No.2(80) P. 219-222 (co-authors Kalibekova A.N., Kasen A.K., doctoral student's share 90%)

10. Sorption capacity of mixed sorbent. Mechanics and Technology / Scientific journal. – 2023.No.4(82). – P.112-115. (co-authors Sembek A., Sayabay A., Begenov A., Serikbayev Sh., Elamanova S.Zh., doctoral student share 90%)

11. Auyr metal cations sorption son sulfokomirdin kyshkyldyk – negizdik ortalyktaryn aseri. XII International Beremzhanov Congress on Chemistry and Chemical Technology, December 4-6, 2024, Almaty, Kazakhstan. Collection of abstracts of reports [Electronic edition]. — Almaty: NJSC "KazNU named after. al-Farabi", 2024. – 222 p. (co-authors Ergimbay S.E., Ryzkiya B.S., doctoral student's share 90%)

Structure and volume of work. The dissertation work consists of standard sections: normative references, designations and abbreviations, introduction, literature review, experimental part, experimental results and their discussion, list of used literature and appendix.

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