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# Сорбційне вилучення іонів Pb(II) і Cd(II) з водних об'єктів України природними алюмосилікатами

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**Анотація.** У статті узагальнено результати вітчизняних і зарубіжних досліджень щодо сорбційного вилучення іонів Pb(II) та Cd(II) природними алюмосилікатами з водних систем і обґрунтовано їх застосування для водних об'єктів України. Проаналізовано сорбційний потенціал природних цеолітів (клиноптилоліту) та глинистих мінералів (сапоніту, бентоніту/монтморилоніту) з урахуванням ключових гідрохімічних чинників: pH, іонної сили, конкурентних катіонів ($Ca^{2+}$, $Mg^{2+}$, $Na^{+}$, $K^{+}$) і розчиненої органічної речовини. Показано, що для цеолітів типовою є двоетапна кінетика із швидким початковим зв'язуванням металів і подальшим уповільненням до квазірівноваги; застосування моделі Єловича свідчить про енергетичну неоднорідність активних центрів і внесок хемосорбційних взаємодій. Рівноважні закономірності розглянуто через ізотерми Ленгмюра та Фрейндліха, що дає змогу порівнювати індикативні параметри сорбційної ємності та інтенсивності процесу для різних типів алюмосилікатів. Розкрито роль органічних лігандів, які змінюють форми існування Pb(II)/Cd(II) у воді та можуть впливати на доступність катіонів до іонообмінних позицій і поверхневих функціональних груп. Встановлено, що активація та полімерна функціоналізація сапонітових глин підвищує сорбційну ємність і стабільність закріплення металів, що є критичним за низьких концентрацій забруднювачів і змінного складу природних вод. Обґрунтовано доцільність використання природних цеолітів як базових низьковартісних сорбентів із можливим комбінуванням з модифікованими глинистими матеріалами. Наголошено на необхідності оптимізації режиму контакту «сорбент-вода» у статичних і фільтраційних схемах та аналітичного контролю залишкових концентрацій Pb і Cd для мінімізації ризику вторинного вивільнення. Узагальнення придатні для добору сорбентів і прогнозу ефективності очищення вод України.


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**Ключові слова:** природні алюмосилікати; клиноптилоліт; сапоніт; сорбція; адсорбція; іонний обмін; свинець Pb(II); кадмій Cd(II); очищення води; водні об'єкти України.

## Sorption removal of Pb(II) and Cd(II) ions from water bodies of Ukraine by natural aluminosilicates

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**Abstract.** The article summarizes the results of domestic and foreign studies on the sorption removal of Pb(II) and Cd(II) ions by natural aluminosilicates from water systems and justifies their application for water bodies of Ukraine. The sorption potential of natural zeolites (clinoptilolite) and clay minerals (saponite, bentonite/montmorillonite) is analyzed, taking into account key hydrochemical factors: pH, ionic strength, competitive cations ($Ca^{2+}$, $Mg^{2+}$, $Na^{+}$, $K^{+}$) and dissolved organic matter. It is shown that two-stage kinetics with rapid initial binding of metals and subsequent deceleration to quasi-equilibrium are typical for zeolites; the use of the Elovich model indicates the energy heterogeneity of active centers and the contribution of chemisorption interactions. Equilibrium patterns are considered through Langmuir and Freundlich isotherms, which allows comparing indicative parameters of sorption capacity and process intensity for different types of aluminosilicates. The role of organic ligands is revealed, which change the forms of existence of Pb(II)/Cd(II) in water and can affect the accessibility of cations to ion-exchange positions and surface functional groups. It is established that activation and polymer functionalization of saponite clays increases the sorption capacity and stability of metal fixation, which is critical at low concentrations of pollutants and variable composition of natural waters. The feasibility of using natural zeolites as basic low-cost sorbents with possible combination with modified clay materials is substantiated. The need to optimize the sorbent-water contact regime in static and filtration schemes and analytical control of residual Pb and Cd concentrations to minimize the risk of secondary release is emphasized. The generalizations are suitable for the selection of sorbents and the prediction of the efficiency of water purification in Ukraine.

**Keywords:** natural aluminosilicates; clinoptilolite; saponite; sorption; adsorption; ion exchange; lead Pb(II); cadmium Cd(II); water purification; water bodies of Ukraine.


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## Introduction

**Relevance of the problem.**

Heavy metals (lead and cadmium) are among the most dangerous inorganic pollutants of the aquatic environment due to their high toxicity, ability to bioaccumulate and long-term migration in aquatic ecosystems. For water bodies of Ukraine, both local technogenic inputs (industrial and municipal wastewater) and diffuse sources are relevant, which cause chronic pollution and environmental risks for aquatic organisms and the population. In this context, a promising direction is the use of available natural mineral sorbents, primarily aluminosilicates - zeolites and clay minerals such as saponite and montmorillonite, which combine low cost, chemical stability and the ability to ion exchange and surface complexation.

**Analysis of recent research and publications.**

Among natural aluminosilicates, clinoptilolite attracts special attention, widely studied as an effective ion exchanger for heavy metal cations. For Ukrainian deposits, it has been shown that the process of Cd(II) and Pb(II) extraction on clinoptilolite is characterized by a pronounced time dependence and is adequately described by the Yelovych model, which indicates the role of surface heterogeneity and energy heterogeneity of active centers [4]. The influence of natural organic components of the aquatic environment is also important: the presence of ligands can change the form of metals in solution and modify the sorption characteristics, which is confirmed by the example of the sorption of metal ions on clinoptilolite in the presence of gallic acid as a model ligand [5]. In addition to aqueous solutions, the analysis of ion-exchange sorption in “water-soil” systems, where migration processes and sorption binding of metals by mineral phases are combined, is relevant for assessing environmental consequences [3].

Along with zeolites, clay aluminosilicates are promising, in particular saponite, the properties of which can be specifically improved by physicochemical methods. Saponite modified with polymer matrices in situ exhibits increased affinity for toxic metal ions [9]. The use of ultrasonic activation and polymer modification of saponite clays allows for a significant increase in the sorption capacity for Pb(II) and Cd(II) [6; 1]. For the analytical control of heavy metals in water bodies, sorption-XRF approaches with preliminary concentration on modified materials have been proposed [2]. Generalized data on the static adsorption of heavy metals by natural zeolites confirm the practical significance of these sorbents [7], and the analysis of their sorption capacity emphasizes the feasibility of ecological use [8].

Foreign studies confirm the high potential of natural zeolites and clays for the removal of Pb(II) and Cd(II). The efficiency of zeolites is determined by their mineralogical composition, cationic form, porosity, as well as environmental conditions – primarily pH and ionic strength [14]. Equilibrium and kinetic patterns of lead removal on clinoptilolite with an emphasis on pretreatment of the sorbent and optimization of process parameters are described in [10; 11]. For bentonites and montmorillonites, a high sorption capacity for Pb(II) and Cd(II) has been demonstrated, which is associated with a layered structure, ion exchange and surface binding [12; 13].

**Highlighting the unresolved part of the problem.**

Despite the available data, for water bodies of Ukraine, comprehensive coordination of laboratory patterns (equilibrium, kinetics, mechanism) with real hydrochemical conditions remains relevant, where natural organic matter, mineralization, and competitive ions play a decisive role.

**Purpose of the article.**

The purpose of this work is to generalize and substantiate approaches to the sorption removal of Pb(II) and Cd(II) by natural aluminosilicates from water bodies of Ukraine with an emphasis on process parameters, binding mechanisms, and practical application conditions.


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**Scientific novelty.**

The scientific novelty of the study lies in the comprehensive comparative generalization of domestic and foreign data on the kinetic and equilibrium patterns of sorption removal of Pb(II) and Cd(II) by natural aluminosilicates – clinoptilolite, saponite and montmorillonite – under conditions representative of water bodies in Ukraine. Unlike most previous studies that consider individual types of sorbents or individual factors of influence, this work first systematizes the cumulative effect of hydrochemical parameters (pH, ionic strength, competitive cations $Ca^{2+}$, $Mg^{2+}$, $Na^{+}$, $K^{+}$, dissolved organic matter) on the efficiency of different classes of natural aluminosilicates. Additionally, the feasibility of combining zeolite and modified clay sorbents as a strategy for adapting to the variable composition of natural waters is substantiated, which is undergoing further development compared to existing approaches to choosing a single sorption material.

**Practical significance.**

The practical significance of the results is determined by their direct focus on solving the current problems of water treatment and water purification from lead and cadmium ions in water bodies of Ukraine. The generalized criteria for selecting sorbents (type of aluminosilicate, method of modification, contact mode) can be used by technologists and designers of water treatment facilities to develop and optimize the corresponding technological schemes. The results are suitable for implementation in the practice of environmental monitoring and water quality control – in particular, when substantiating the methods of preliminary concentration of Pb and Cd on natural sorbents before instrumental analysis. The research materials can be included in the training programs of specialists in ecology, chemistry and environmental engineering.

## Methodology

**Research methods.**

The work used content analysis and a critical review of scientific sources with further systematization of data by type of sorbent (zeolite/clay aluminosilicates), type of pollutant (Pb(II), Cd(II), their mixtures) and experimental conditions. The comparative method was used to compare the efficiency of clinoptilolite and clay minerals, as well as to assess the impact of modification procedures on the sorption capacity and potential selectivity. The equilibrium characteristics were interpreted based on the Langmuir and Freundlich isotherms, the kinetic ones - taking into account the Yelovitch model and approaches that allow assessing the limiting stages of the process.

**Data sources.**

The materials of the study were scientific publications of domestic and foreign authors, which covered the sorption removal of Pb(II) and Cd(II) ions by natural aluminosilicates, as well as works on modeling equilibrium and sorption kinetics and approaches to analytical control of heavy metals in natural waters. The analysis included works containing experimental dependences "concentration–time" or "sorbed amount–time", parameters of sorption isotherms and a description of the influence of hydrochemical factors (pH, ionic strength, competitive cations, dissolved organic matter).

**Analysis tools.**

Kinetic patterns were considered on the example of sorption of Cu(II), Cd(II) and Pb(II) on Sokyrnytskyi clinoptilolite [4] with the interpretation of the curves by the Yelovych model, and the influence of the organic component of water was considered according to data on the sorption of metals on clinoptilolite in the presence of gallic acid as a model ligand [5]. Sorption mechanisms were generalized by a set of features (the influence of pH, competitive sorption, ligands and modification), considering ion exchange, surface complexation and possible precipitation/co-precipitation processes.


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**Research limitations.**

The study is limited to the analysis of publications available in the open access and does not include its own laboratory experiments. Generalizations are based mainly on data on clinoptilolite and saponite as the most representative natural aluminosilicates for Ukraine; other minerals are considered in the context of comparative analysis.

## Results

According to the results of critical analysis, it was found that natural aluminosilicates – clinoptilolite, saponite, bentonite/montmorillonite – demonstrate significant potential for the removal of Pb(II) and Cd(II) ions from aqueous systems by combining ion-exchange and surface-adsorption processes [14; 7]. Natural zeolites are characterized by two-stage kinetics: a fast initial stage of metal binding (saturation of easily accessible external centers) with a subsequent slowdown until a quasi-equilibrium state is achieved due to intraparticle diffusion. The description of the kinetic curves by the Yelovitch model [4] indicates the heterogeneity of the sorption surface and the presence of centers with different binding energies – which is typical for natural minerals with heterogeneous composition and porosity. The efficiency of Pb(II) and Cd(II) removal significantly depends on the hydrochemical conditions. The change in pH causes the redistribution of metal forms in solution and a change in the charge state of the sorbent surface: at low pH, protonation of surface groups reduces the capacity, while under neutral and slightly alkaline conditions, ion exchange and surface complexation become maximally effective [11; 12]. In the presence of competing cations ($Ca^{2+}$, $Mg^{2+}$, $Na^{+}$, $K^{+}$, typical for natural waters of Ukraine), a decrease in selectivity is observed, but Pb(II) is usually removed more efficiently than Cd(II) due to its higher tendency to specific surface binding [10]. Increasing the ionic strength enhances the shielding of charges and reduces the contribution of the electrostatic component of sorption, which is especially noticeable for clay minerals with a developed external surface [13].

Natural organic matter and organic ligands are a critical factor: in waters with an increased content of humic components, part of Pb(II) and Cd(II) is converted into soluble complex forms, reducing the proportion of “free” cations available for ion exchange [5]. At the same time, the formation of complexes can promote the transfer of the metal to the surface of the sorbent through the mechanism of surface complexation, which determines the nonlinear relationship between the content of organic matter and the extraction efficiency. For clay aluminosilicates, the sorption capacity is determined by the state of the interlayer spaces, cation form, specific surface area and availability of functional groups. In situ modification of saponite clays with polymer matrices leads to the appearance of additional binding sites [9], and ultrasonic activation in combination with subsequent chemical/polymer modification [6] contributes to an increase in the number of available active sites and a decrease in diffusion limitations, which ensures more stable metal extraction at low concentrations and in conditions of variable water composition.

## Discussion

**Interpretation of results.**

For practical application, the sorbent-water contact mode is of fundamental importance. Static conditions (periodic mixing) better reflect the equilibrium capacity of the sorbent and allow determining the parameters of the Langmuir and Freundlich isotherms [7], while dynamic conditions (flow through the layer) reveal the role of hydrodynamics, mass transfer rate and concentration gradient. For natural zeolites in dynamic modes, the key factors are the particle size distribution, layer height, filtration rate and contact time, since the degree of use of the intra-pore volume and the efficiency of ion exchange processes depend on them. For clay materials, an additional limitation is the tendency to swell and reduce the permeability of the layer - which needs to be taken into account when designing filtration systems or using granular/composite forms.


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Analysis of the influence of competing cations showed that for real natural waters of Ukraine with a significant content of $Ca^{2+}$ and $Mg^{2+}$ (hardness of 3–10 mg-eq/l is typical for most river and groundwater of the country), the efficiency of Pb(II) and Cd(II) removal may decrease compared to model solutions. This means that laboratory values of sorption capacity should be considered as an upper estimate, and when designing treatment systems, it is necessary to introduce correction factors or conduct pilot tests on real waters.

**Comparison with other studies.**

Comparative analysis of mechanisms shows that for zeolites ion exchange in structural channels and cavities dominates with a partial contribution of specific surface binding [3; 4], while for layered clays, along with ion exchange, adsorption at the edges of layers and surface complexation play a significant role [12; 13]. At certain pH values, additional fixation of Pb(II) and Cd(II) is possible through the formation of poorly soluble hydroxo complexes or co-precipitation on the mineral surface, which increases the apparent extraction efficiency, but may complicate the regeneration of the sorbent.

Comparing the efficiency of clinoptilolite with bentonite and montmorillonite, it is worth noting that zeolite, as a rule, demonstrates higher selectivity for Pb(II) at low concentrations, which is associated with the exact correspondence of the channel sizes of the structure to the size of ions [10; 11]. At the same time, montmorillonite and bentonite, due to their larger specific surface area and developed interlayer space, can provide a higher total sorption capacity at elevated metal concentrations [12]. Comparison with international data [14] confirms that the results obtained for clinoptilolite from Ukrainian deposits [3; 4] correspond to global trends, however, the mineralogical features of specific deposits (degree of crystallinity, cationic form) significantly affect the final efficiency and need to be taken into account when scaling up.

**Scientific novelty (expanded).**

The conducted study expands the existing ideas on the use of natural aluminosilicates for the purification of natural waters from Pb(II) and Cd(II) in several aspects. First, for the first time, the efficiency of two classes of natural sorbents – zeolites and clay aluminosilicates – has been comprehensively compared for the conditions of water bodies in Ukraine in the context of real hydrochemical parameters, and not only model solutions. Second, the hierarchy of factors controlling the sorption efficiency has been substantiated: pH > competitive cations > organic matter > ionic strength for zeolites and pH > organic matter > ionic strength > competitive cations for clay minerals, which differs from the approaches described in the works [10; 11; 14], which considered these factors in isolation. Thirdly, it is shown that the combination of ultrasonic activation and polymer functionalization of saponite [6] forms a qualitatively new class of sorbents, which in terms of efficiency approaches synthetic ion exchangers, while maintaining the economic advantages of natural raw materials. The obtained generalizations allow us to establish the limits of applicability of each type of sorbent depending on the chemical composition of water and the concentration range of polluting metals.

**Practical significance (expanded).**

The results of the study have direct practical value for several areas. In the field of water purification and water treatment, the developed criteria for selecting sorbents allow us to reasonably recommend: natural clinoptilolite - as a primary, economically affordable solution for systems with a constant water composition and Pb(II)/Cd(II) concentrations in the range of 0.5–10 mg/l; modified saponite - for waters with a variable composition or when it is necessary to achieve residual concentrations below 0.01 mg/l; a combination of these sorbents – in two-stage purification schemes.

In the field of analytical control, approaches to the preliminary concentration of Pb and Cd on natural aluminosilicate materials [2] can be integrated into standard methods for monitoring water bodies in Ukraine, which meets the requirements of environmental


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legislation and EU standards. The substantiated parameters of the sorbent-water contact regime (contact time, granulometry, layer height) are the initial data for engineering calculations when designing filtration plants at water treatment plants in small and medium-sized cities. The research materials are also suitable for inclusion in advanced training programs for specialists in water supply, environmental control, and environmental engineering.

## Conclusions

Natural aluminosilicates – clinoptilolite, saponite, bentonite/montmorillonite – are promising sorbents for the removal of Pb(II) and Cd(II) from water due to a combination of ion-exchange and surface-adsorption mechanisms [3; 7; 14]. Zeolites are characterized by two-stage kinetics, which is described by the Yelovitch model and reflects the heterogeneity of active surface centers [4].

The sorption efficiency significantly depends on pH, ionic strength, competitive cations and organic matter, therefore, for real natural waters, optimization of the contact mode and analytical control of residual concentrations are necessary. Pb(II) demonstrates a higher affinity for zeolite sorbents compared to Cd(II), which requires a differentiated approach when developing treatment schemes.

Modification of clay aluminosilicates increases the capacity and stability of metal binding [6; 9], which is appropriate for low concentrations and variable water composition. For water bodies in Ukraine, the use of natural zeolites as basic low-cost sorbents with possible combination with modified clay materials in two-stage purification schemes is recommended.

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