ЕКОЛОГІЧНА БЕЗПЕКА, ОХОРОНА ПРАЦІ

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APPLICATION OF NATURAL SORBENTS FOR WASTE DETOXICATION

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ЗАСТОСУВАННЯ ПРИРОДНИХ СОРБЕНТІВ ДЛЯ ДЕТОКСИКАЦІЇ ВІДХОДІВ

Purpose. To study the possibility of use of absorption properties of natural sorbent agents for waste detoxication.

Methodology. To study the chemical composition of wastes, sediments and in laboratory modeling of waste detoxication we applied the particle-size, bulk and atomic absorption spectrophotometric methods of analysis. For processing the results of the research, the methods of mathematical statistics were used.

Findings. The products of solid waste combustion contain 92–97% of heavy metals of danger class 1 and 3–8% of metals of danger class 2. The content of mobile fractions of heavy metals in incineration products formed the following descending series: for ash – Zn>Pb>Cr>Cd>Cu>Ni>Co; for slag – Zn>Pb>Cu>Cr>Ni>Cd>Co. The influence of the rocks sorbents absorption properties, chemical properties of heavy metals and geochemical form of metal on the effectiveness of waste detoxication was determined. To detoxify the waste, the carbonate clay with the mass fraction of 30% is recommended to be used. The detoxication of waste occurs due to the absorption properties of natural sorbents and as a result of the dilution of waste by neutral natural materials.

Originality. It is shown that the processes of sorption immobilization lead to a significant decrease in the content of mobile fractions of heavy metals in the waste and thus help to reduce its environmental hazard. It was determined that the heavy metals of danger class 1 better undergo transformation into immobile forms than the heavy metals of the danger class 2. To evaluate the efficiency of detoxication of wastes by natural sorbents, it was proposed to use the detoxication rate.

Practical value. The ability of natural sorbents to turn heavy metals into low-solubility compounds reduces their migratory activity. The use of natural sorbents for detoxication of waste will increase the level of environmental safety of biological systems and human health.

Keywords: *waste, heavy metals, natural sorbents, detoxication, environmental safety*

The problem substantiation. As a result of large volumes of industrial and municipal wastes the problem of their utilization becomes very urgent in Ukraine. The densely populated and industrialized Dnipropetrovsk region is one of the leaders according to volumes of industrial and municipal wastes as more than 9 milliard tones of wastes (64,6% of the total country volume) are stored on its territory [1].

Ukraine lacks for landfills for industrial and municipal wastes. The wastes occupying large territories and often stored in a way not permitted by environmental regulations are especially dangerous for environment and living organisms. The uncontrolled disposal of large quantities of waste causes the contamination of residential areas in population aggregates and adjoining agricultural lands with heavy metals (HM) [2–5].

Incineration is a waste treatment method for municipal solid waste (MSW), which enables wastes volume reduction and the reduction of landfills area. MSW combustion causes the concentration of HM and other toxic components in combustion products. It should be pointed out that the products have potential toxic and mutagenic properties and negatively affect the condition of ecosystems components and population health [6].

The need for timely processing and utilization of industrial and municipal wastes containing HM is one of the most important problems of hydroecology and environment protection.

Review of the related researches. The use of unconventional, widespread and cheap natural materials is of great practical interest for the solution of the problem. The latest scientific researches are focused on the application of mineral silica-alumina sorbents (clays, zeolites and other materials with good absorption properties and environment resistance) to immobilize HM. The special attention is paid to the application of montmorillonite clays for HM immobilization. The crystal structure peculiarities and surface chemistry of the minerals specify their great adsorption properties [7]. Many scientific papers provide the compositional (chemical and mineralogical) analysis of montmorillonite clays and describe their colloid-chemical properties and the ability of the naturally occurring, concentrated and modified clay forms to absorb the HM ions; demonstrate that the clays from different deposits excel such effective sorbents as wood charcoal and activated carbon in their sorption properties [3,7].

The possibilities to apply clays in the detoxication of MSW combustion products (HM removal) are insufficiently explored, but it is considered to be a prospective and productive way of the urgent ecological problems solving [8, 9].

The identification of unresolved issues. The problem of waste combustion products storage and disposal consists of the urgent issues to be solved to provide the sustainable development of the state. They are the reliable estimate of wastes toxicity level, their detoxication and recycling. According to the geochemical theory the toxicity of waste combustion products containing HM is closely connected with the forms of existence in the environment. HM contaminate the environment only by means of their mobile fractions, which can migrate to the environment and then move by trophic lines.

The solution of the raised problem is based on the concept of binding of HM mobile fractions in waste combustion products by natural dispersed materials.

The purpose formulation. The wastes toxicity level reduction in case of natural sorbent agents application is caused by two factors: firstly, as a result of the display of natural dispersed material absorption properties, the mobile fractions of HM are transformed into immobile ones, the toxic elements are converted from soluble state into low-solubility compounds; secondly, because of the dilution of hazardous waste combustion products by neutral environmentally-friendly natural material, the abundance ratio of HM in the final product i.e compound is reduced. So, the research into natural sorbent agents application for reduction of wastes toxicity (caused by HM) is of great importance.

The Purpose of the work is the investigation of the application potential of absorption properties of natural sorbent agents in wastes detoxication.

The main part. The object of the research is the ash and slag produced by Dnipropetrovsk Waste Incineration Plant (DIP). The combustion wastes toxicity was estimated with respect to mobile fractions of HM of danger classes 1, 2: Pb, Cd, Zn, Co, Ni, Cu, Cr. The mobile fractions of HM were separated from the wastes by acetate and ammonium buffer with pH = 4.8. The metals content in wastes before and after detoxication was estimated by the atomic absorption spectrometry method.

The content of mobile fraction of HM in the ash and slag was high. Thus, almost 80% of plumbum compounds and 100% of zink compounds passed from slag to buffer solution, 65% of plumbum compounds, 49% of zink compounds, 100% of nickel compounds, and 63% of cobalt compounds were separated from ash.

It was established that both ash and slag are characterised by the highest content of mobile fractions of HM of the danger class 1: zink (3400 in ash and 1400mg/kg in slag) and plumbum (650 and 240mg/kg respectively). It should be noted that the content of cadmium (56mg/kg) and chromium (60mg/kg) in the ash was high as well as the content of copper (115mg/kg) in slag. The amount of nickel and cobalt did not exceed 20mg/kg.

According to the content of HM mobile fraction in DIP waste combustion products the metals could be ranged in descending order as follows:

- for ash – *Zn>Pb> Cr>Cd>Cu>Ni>Co*;
- for slag – *Zn>Pb>Cu>Cr>Ni>Cd>Co*.

It was determined that the percentage of HM of danger class 1 in the general content of mobile fraction is 97% for ash and 92% for slag. The percentage of the metals of danger class 2 is 3% for ash and 8% for slag. Thus, the data obtained prove that the investigated wastes constitute a serious threat for environment and must be detoxified.

The experimental investigation of DIP ash and slag detoxication (mobile fraction of HM remove) was conducted with the application of such natural sorbent agents as clays.

It is known that the main conditioning factors for absorption properties of natural sorbent agents are carbonate minerals content and cation exchange capacity (CEC). So, in this study, it was planned to determine the combined influence of all the factors mentioned on the efficiency of the detoxication of wastes containing HM.

The clays with general carbonate minerals content of 16.6% (carbonate clay, Sample 1) and 1.4% (noncarbonated clay, Sample №2) as well as clays with CEC rate of 30.3mg-equiv/100g (Sample 1) and 38.4mg-equiv/100g (Sample 2) were sampled in Dnipropetrovsk region.

Sample 1 is the brown colour clay and Sample 2 is the green and yellow-grey clay. Sample 1 has liquid inclusions and white colour ties represented by calcite. The inclusions are from 1 to 4mm in size. The clay (Sample 2) is homogeneous, without inclusions and ties. The clays are hard, soapy and stratified. The surface of a lump sample is smooth; the frace has platy or foliation parting. The studied clays are of hygroscopic type, imbibe water rapidly without swelling. The clays have gel (Sample 1) and silty (Sample 2) microstructures. The aggregates of clay particles are of light-yellow and ochraceous colours. The interference tints are light-yellow with light-gray sections, which is indicative of caolinite and hydromicaceous composition (Specimen 1). The interference tints in the rock of the Specimen 1 are yellow and spice orange, which indicates the hydromicaceous composition. The inclusions are from 1 to 4mm in size. The clay particles are between 0.005 and 0.01mm in size. The physical specifications of rock samples are tabulated in table 1.

Table 1

The physical specifications of rock samples

Index		Carbonate clay	Non-carbonated clay
Humidity, %		7.62	6.72
Density g/cm^3		2.73	2.76
Particle-size composition, %	clay	31.70	44.00
	siltstone	66.60	54.20
	sand	1.70	1.20

The analysis of the data from table 1 demonstrated that clay fraction percentage in the samples studied is from 31.7 to 44%. The clay samples contain clay particles in the form of sporadical fission units of aleurite fraction as well as plaster-stone, sodium-calcium feldspar and ferric hydroxides. The rock contains aleurite fraction (up to 66.6 %) with fraction grain size from 0.005 to 0.05mm, some of the grains are 0.15mm in size. The sandy fraction with grain size up to 0.2mm is present too. The percentage of this fraction is 1.2–1.7%. The sandy fraction is represented by plaster-stone, quartz, anhydrite and feldspars. The rock structure is of argillaceous texture type.

The results of the study of salts content in the aqueous extracts of sorbent rocks are tabulated in table 2.

To provide more comprehensive estimation of the rocks under study, the content of mobile fraction of HM in the rocks was determined (table 3).

Table 2

The results of the analysis of sorbent rocks aqueous extracts

Index		Carbonate clay	Non-carbonated clay
HCO_3^-	mg/g	1.22	0.73
	%	0.12	0.07
SO_4^-	mg/g	0.24	0.38
	%	0.02	0.04
Cl^-	mg/g	0.75	0.25
	%	0.08	0.03
Ca^{2+}	mg/g	0.06	0.08
	%	0.01	0.01
Mg^{2+}	mg/g	0.05	0.07
	%	0.01	0.01
$Na^+ + K^+$	mg/g	0.98	0.39
	%	0.10	0.04
Total	mg/g	2.64	1.54
	%	0.26	0.15

Table 3

The content of mobile fraction of HM in the rocks, mg/kg

Clay type	Pb	Cu	Cd	Zn	Mn	Ni	Co	Cr
Carbonate	8.0	2.0	1.5	2.0	75.0	3.0	2.0	5.0
Non-carbonated	6.0	0.0	0.0	0.5	6.0	2.0	0.4	0.0

The results of analysis of rocks chemical composition demonstrated that the salinity rate and content of mobile fraction of HM are small. Later the content of mobile fraction of HM was taken into consideration in the calculations of the wastes detoxication degree with clays application.

The wastes detoxication technique was as following: the definite proportions of dry ash (slag) and clay were mixed up. Then the 10 dm^3 of distilled water were added, the mixture was blended to provide watering and the samples were put aside until complete drying. The dried cemented matter was crushed and triturated, then the content of mobile fraction of HM was determined using the standard methods described by Je.V. Arinushkina.

The efficiency of the detoxication process of MSW combustion products was determined according to the content of mobile fraction of HM in the wastes after their interaction with a natural sorbent agent and the detoxication degree.

The wastes detoxication degree was determined with respect to both clay absorption properties and the dilution of hazardous MSW combustion products by natural material, formula evaluation is

$$\omega = \frac{C_0 - C_K}{C_0},$$

where ω is the detoxication degree; C_0 is HM content in ash (slag) before the detoxication, mg/kg; C_k is HM content in the mixture after the detoxication, mg/kg.

To calculate the optimum quantity of the sorbent agent to be used for wastes detoxication, the sorbent mass and detoxication degree ratio was studied. For the unchanged mixture mass the percentage of the clay used to provide the wastes detoxication was 10 and 30%. The fig. 1–4 demonstrate the results of the researches.

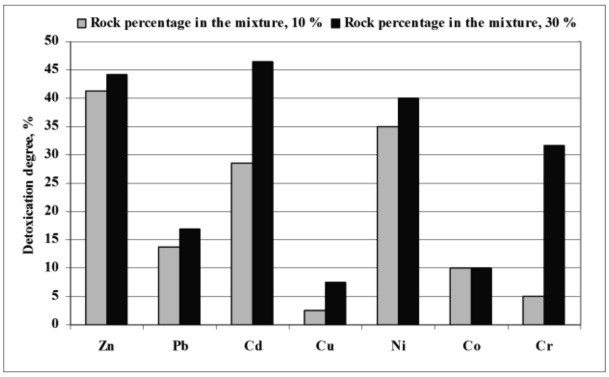

Fig. 1. The ash detoxication degree provided by carbonate clay

The data analysis based on the fig. 1 demonstrates that in case of ash detoxication provided by carbonated clay the transformation process of mobile fractions of all studied metals takes place. The increase of the sorbent clay percentage causes the increase of ash detoxication degree (almost all mobile fractions of HM are removed). Cobalt is an exception. The slag detoxication degree related to this metal does not depend on the percentage of carbonate clay.

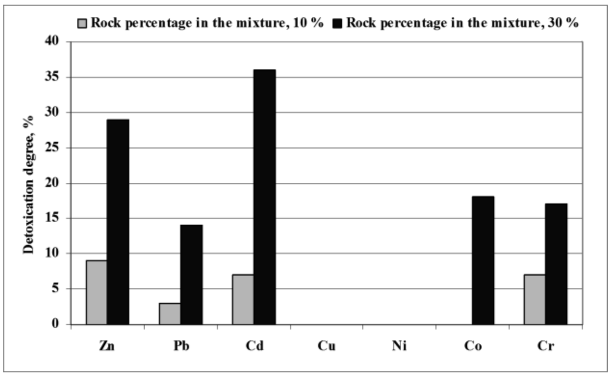

Fig. 2. The ash detoxication degree provided by non-carbonated clay

The data analysis based on the fig. 2 demonstrates that the decrease in content of HM mobile fractions in case of ash detoxication provided by non-carbonated clays (Sample 2) takes place only for five metals. Copper and nickel are not detoxified in the ash studied. The clay percentage increase provides the tangible detoxication degree increase (from 2.5 to 5 times).

The same regularities are observed in case of slag detoxication provided by clays (fig. 3, 4). The carbonate clay has better binding properties for mobile fractions of HM. The application of 10% of clay for slag detoxication provides the decrease of mobile fractions content for only five HM. The carbonate clay does not provide the transformation of cadmium and nickel, the non-carbonated clay is ineffective with cadmium and plumbum. If the percentage of clay used for detoxication is increased up to 30% the change of mobile fraction content of all metals under study is observed. It should be pointed out that the value of slag detoxication degree demonstrates the 2–4 times increase in case of carbonate clay application; in case of non-carbonated clay the value does not go up, with the exception of cadmium and plumbum.

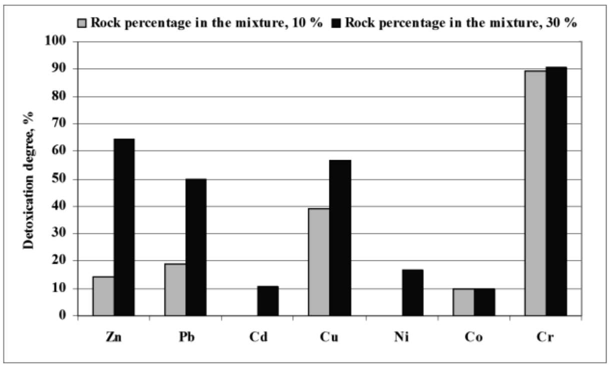

Fig. 3. The slag detoxication degree provided by carbonate clay

According to the ability of mobile fractions of HM to transform into immobile ones as result of the detoxication process of waste combustion products in case of clay application, HM could be ranged as follows:

Carbonate clay:

for ash : *Cd>Ni>Zn>Cr>Pb>Cu>Co;*

for slag : *Cr>Zn>Cu>Pb>Ni>Cd>Co*.

Non-carbonated clay:

for ash : *Cd>Ni>Zn>Cr>Pb>Cu>Co*;

for slag : *Cr>Zn>Cu>Pb>Ni>Cd>Co*.

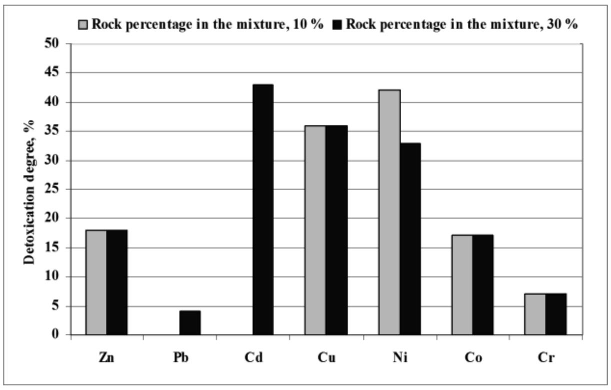

Fig. 4. The slag detoxication degree provided by non-carbonated clay

The fig. 5 provides the data of the total quantity of mobile fractions of HM transformed into immobile ones as a result of the detoxication process of waste combustion products when applying clays with different carbonate content.

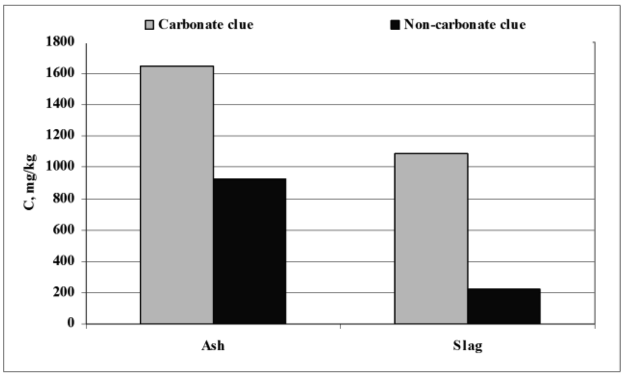

Fig. 5. Total content of HM being transformed into immobile fractions

It is proposed to estimate the efficiency of the detoxication process of waste combustion products in case of clays application according to the detoxication ratio. The detoxication ratio formula evaluation is

$$K_d = \frac{\sum C_0}{\sum C_K},$$

where K_d – waste detoxication ratio; C_0 – HM content before the waste detoxication, mg/kg; C_k – HM content after the waste detoxication, mg/kg.

The detoxication rate value describes how the total quantity of HM decreased during the wastes detoxication process. Tables 4 and 5 show detoxication rates for different variants of waste combustion products neutralization with clays application.

Table 4

Detoxication rate values for waste incineration ash

Clay type	Detoxication rate value		
	K_d	K_d (I)	K_d (II)
Carbonate	1.65	1.64	1.32
Non-carbonated	1.29	1.29	1.09

Remarks: K_d is the general wastes detoxication rate; K_d (I) is the wastes detoxication rate for metals of danger class 1; K_d (II) is the wastes detoxication rate for metals of danger class 2.

Table 5

Detoxication rate values for waste incineration slag

Clay type	Detoxication rate value		
	K_d	K_d (I)	K_d (II)
Carbonate	2.59	2.63	2.18
Non-carbonated	1.16	1.14	1.48

The data from tables 4, 5 demonstrate that the change of the degree of the content of HM mobile fractions is higher for slag detoxication with the studied clays application than for ash detoxication. HM of danger class 1 better undergo transformation into immobile forms than metals of danger class 2. The reason is that the HM of danger class 1 prevail in the total quantity of the HM studied in wastes.

The comparative analysis of the ash and slag detoxication processes (mobile fractions of HM removal) indicated that the transformation into immobile forms in slag is more complete. The fact is reasoned firstly by the smaller initial content of mobile fractions of all studied HM in slag compared to ash. Secondly, we should admit that the aqueous extract of slag has a higher value of acid-base balance (pH =11.55) compared to the aqueous extract of ash (pH =10.20). This factor affects the detoxication degree of wastes combustion products as the studied processes occur at the boundary of liquid and solid phases. Thus the chemical properties of the slag provide better conditions for absorbing mobile fractions of HM.

Thereby, the main factors affecting the wastes detoxication degree in case of mobile fractions of HM neutralization are as following:

- the absorption properties of sorbent agents;
- the chemical properties of metal;
- the geochemical fraction of metal in wastes;
- qualitative and qualitative relation of HM in wastes.

Conclusions and further research. As a results of investigations conducted it was found that:

- disperse materials and sediments are recommended to be used for the detoxication of such complicated multicomponent systems as MSW combustion;
- the immobilization degree of HM in wastes increases with the increase of total content of each of them and it is limited by the value of absorption capacity of the sorbent agent;
- in case of combined presence of 3–5 HM in wastes the selective adsorption in the process of their immobilization by sediments is observed;
- carbonate clay provides the maximum degree of neutralization of HM mobile fractions in ash and slag detoxication;
- the detoxication process is of the maximum effectiveness if the mass fraction of carbonate clay is 30%.

Thus, as a result of the study it was proved that the disperse materials of aluminosilicate type have high absorption capacity concerning HM in the MSW combustion products. It should be pointed out that different rocks vary according to their absorption capacities concerning the definite metals. So, the sorbent materials should be chosen with respect to chemical composition of the wastes to be neutralized.

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Мета. Дослідження можливості використання поглинальних властивостей природних сорбентів для детоксикації відходів.

Методика. Для вивчення хімічного складу відходів, осадових порід та при проведенні лабораторного моделювання процесів детоксикації відходів застосовані гранулометричний, об'ємний та атомно-абсорбційний спектрофотометричний методи аналізу. Для обробки результатів досліджень використані методи математичної статистики.

Результати. Продукти термічної переробки твердих побутових відходів містять 92–97% важких металів I класу небезпеки та 3–8% металів II класу небезпеки. За вмістом рухомих форм важких металів у відходах сміттєспалювання в порядку зменшення утворюються наступні ряди: для золи – *Zn>Pb>Cr>Cd>Cu>Ni>Co*; для шлаку – *Zn>Pb>Cu>Cr>Ni>Cd>Co*. Визначено вплив поглинальних властивостей порід-сорбентів, хімічних властивостей важких металів та геохімічної форми металу на ефективність детоксикації відходів. Для детоксикації відходів рекомендується використовувати карбонатну глину з масовою часткою 30%. Детоксикація відходів відбувається завдяки поглинальним властивостям природних сорбентів та в результаті розведення відходів нейтральними природними матеріалами.

Наукова новизна. Доведено, що процеси сорбційної іммобілізації призводять до суттєвого зниження вмісту рухомих форм важких металів у відходах і тим самим сприяють зниженню їх екологічної небезпеки. Встановлено, що важкі метали I класу небезпеки краще зв'язуються в нерухомі форми, ніж метали II класу небезпеки. Для оцінки ефективності процесу детоксикації відходів природними сорбентами запропоновано використовувати показник – коефіцієнт детоксикації.

Практична значимість. Здатність природних сорбентів переводити важкі метали у важкорозчинні сполуки дозволяє знизити їх міграційну активність. Використання природних сорбентів для детоксикації відходів дозволить підвищити рівень їх екологічної безпеки для біологічних систем та здоров'я людини.

Ключові слова: *відходи, важкі метали, природні сорбенти, детоксикація, екологічна безпека*

Цель. Исследование возможности использования поглотительных свойств природных сорбентов для детоксикации отходов.

Методика. Для изучения химического состава отходов, осадочных пород и при проведении лабораторного моделирования процессов детоксикации отходов применены гранулометрический, объемный и атомно-абсорбционный спектрофотометрический методы анализа. Для обработки результатов исследований использованы методы математической статистики.

Результаты. Продукты термической переработки твердых бытовых отходов содержат 92–97% тяжелых металлов I класса опасности и 3–8% металлов II класса опасности. По содержанию подвижных форм тяжелых металлов в отходах мусоросжигания в порядке уменьшения образуются следующие ряды: для золы – *Zn>Pb>Cr>Cd>Cu>Ni>Co*, для шлака – *Zn>Pb>Cu> Cr>Ni> Cd>Co*. Определено влияние поглотительных свойств пород-сорбентов, химических свойств тяжелых металлов и геохимической формы металла на эффективность детоксикации отходов. Для детоксикации отходов рекомендуется использовать карбонатную глину массовой долей 30%. Детоксикация отходов происходит благодаря поглощающим свойствам природных сорбентов и в результате разведения отходов нейтральными природными материалами.

Научная новизна. Доказано, что процессы сорбционной иммобилизации приводят к существенному снижению содержания подвижных форм тяжелых металлов в отходах и тем самым способствуют снижению их опасности. Установлено, что тяжелые металлы I класса опасности лучше связываются в неподвижные формы, чем металлы II класса опасности. Для оценки эффективности процесса детоксикации отходов природными сорбентами предложено использовать показатель – коэффициент детоксикации.

Практическая значимость. Способность природных сорбентов переводить тяжелые металлы в труднорастворимые соединения позволяет снизить их миграционную активность. Использование природных сорбентов для детоксикации отходов позволит повысить уровень их экологической безопасности для биологических систем и здоровья человека.

Ключевые слова: *отходы, тяжелые металлы, природные сорбенты, детоксикация, экологическая безопасность*

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