

METAL SORPTION IN MODEL WATERS AND WASTEWATER ON ACTIVATED CARBON ADSORBENTS FROM ALMONDS AND WALNUTS.

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Article Received: 27 August 2026, Article Revised: 17 September 2026, Published on: 07 October 2026

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Doi: <https://doi-doi.org/101555/ijarp.3801>

ABSTRACT

The scientific significance of the data presented in this article lies in the fact that they form the basis for the development of a technology for producing activated carbon adsorbents by modifying walnut and almond shells and using the resulting adsorbent for the purification of metals and wastewater from metallurgical plants. The practical significance of these results lies in the development of an industrial technology for producing activated carbon adsorbents from local agricultural waste and their use in industrial wastewater treatment.

KEYWORDS: *Activated carbon, adsorption, metals, surface area, porous structure, environmental safety, industrial waste, treatment processes.*

Currently, coal raw materials are predominantly directed toward the production of solid fuels and are considered one of the main energy sources of the Republic. Today, coal is used for the production of fuel, new reagents, and new materials. More than 5,000 products are manufactured on the basis of more than 130 chemical semi-finished products produced. In recent years, fundamental climate change has necessitated the use of alternative energy

sources. Therefore, approximately 70% of electricity is generated through the use of coal [1,2; pp. 581–585].

The development of technological foundations for solving environmental problems associated with the accumulation of harmful substances in the environment has become an important issue, particularly in terms of rational and efficient use of available opportunities and resources while focusing on achieving the desired results.

Heavy metals, oil, and petroleum products are among the main pollutants of water resources. During oil refining processes, residues or products of petroleum may enter water bodies, resulting in global water pollution. Taking these factors into consideration, the development and production of highly efficient sorption materials is of considerable importance. To date, more than 300 sorption materials have been developed, differing in their methods of preparation, activation, and modification. The production of sorption materials based on agricultural wastes for removing petroleum products from water is considered economically and technologically justified. Two approaches have been applied to improve water purification efficiency and the properties of sorbents: modification of sorbents to improve their sorption characteristics and the use of natural sorption materials to obtain new composites. The preparation of biosorbents with improved properties has also been investigated in [2; pp. 111–116].

In recent years, increasing attention has been paid to carbon-containing products obtained from plant wastes, as well as to wood-processing wastes, which possess specific properties and are suitable for thermochemical activation processes [3; p. 105].

Both granular and powdered activated carbon can be used for liquid purification. Granular activated carbon is mainly used in continuous processes, whereas powdered activated carbon is more suitable for batch processes. The simplest method for separating an adsorbent from a liquid phase is sedimentation; however, it is rarely used because of the long duration of the process. This method is generally used for the purification of wine and other alcoholic beverages.

Activated carbon is widely used in the deodorization of fats and oils. Fine-pore activated carbon is used for these purposes. Granular activated carbon is used only in special cases, for example, for the deodorization of high-molecular-weight oils. Many chemical substances contain large amounts of coloring impurities. These impurities can be removed by treatment with activated carbon. Activated carbon is more frequently used for the purification of natural organic compounds than inorganic substances. Despite improvements in distillation

technologies, glycerin formed during the lipolysis process in soap production still requires treatment with activated carbon, particularly at the final stage. A high degree of purification is required in the production of medical and pharmaceutical preparations, food products, and cosmetics; therefore, powdered activated carbon is widely used [4; pp. 120–132]. Adsorbents play an important role in industrial wastewater treatment, rational utilization of natural resources, and preservation of the biosphere. Therefore, carbon-based adsorbents are widely used to remove various harmful substances from the hydrosphere. Currently, inorganic sorption materials are also widely used for the removal of hazardous substances. Consequently, the synthesis or production of sorbents with targeted sorption properties is of great importance [5; pp. 35–40].

In addition, adsorbents can be obtained from conventional and unconventional types of raw materials for the purification of aqueous and gaseous systems. Brown coal is considered an inexpensive and readily available raw material for the production of adsorbents. The possibility of obtaining carbon-based adsorbents from brown coal has been demonstrated. The resulting materials are characterized by microporous structures and high adsorption capacity. One of the most effective activation methods is alkaline-thermal activation at 600–900 °C.

As a continuation of this research, chemical modification of brown coal using alkali was proposed, resulting in the preparation of activated carbon samples with particle sizes of >0.5–1.0 and >1.0–2.0 mm [6; pp. 31–34].

Coal powder with a particle size of 0.2 mm was mixed with potassium hydroxide at a ratio of 1:1 and held for 60 minutes, followed by thermolysis at 800 °C. Regardless of the type of coal, this method makes it possible to obtain an adsorbent with a high sorption capacity and an iodine adsorption value of up to 96%. At the same time, the adsorbent yield is approximately 50%, while the amount of losses is about 40%. Therefore, natural sorbents are advantageous due to their low cost and availability, as well as the possibility of using them once without regeneration. The authors of [7; pp. 27–32] investigated the possibility of obtaining adsorbents from various fruits and seeds. Valuable products were first separated from these materials, after which they were carbonized at 250–300 °C. Activation was subsequently carried out using additional oxidizing gases and water vapor, and their sorption properties were determined. It should be noted that the resulting adsorbents did not possess a well-developed porous structure. However, since walnut shell materials have a structured surface, they can potentially be used as filters for the removal of high-molecular-weight compounds. A large amount of water is used in the metallurgical industry, resulting in the

generation of significant quantities of wastewater. Since wastewater contains organic substances and residues of various metals, it cannot be discharged directly into sewage systems and is therefore stored in large reservoirs. Due to the lack of wastewater treatment technologies in the Republic, these wastes can cause significant environmental damage. Furthermore, the presence of valuable, rare, or radioactive substances in such wastewater creates opportunities for their recovery and for the reuse of treated wastewater as technical water. For this reason, many researchers are conducting scientific investigations aimed at developing methods for separating these substances and enabling the subsequent reuse of wastewater [8; pp. 29–33].

Variations in the concentrations of metal ions in industrial wastewater also lead to changes in the amount of adsorbent required during the purification process. To determine the sorption capacity of the adsorbent, model solutions containing different concentrations of metal ions were treated using different amounts of the adsorbent, and the adsorption process was investigated.

Table 1: Mass spectrometry results of metallurgical industry wastewater before and after treatment.

Element	Metallurgical wastewater before treatment	Metallurgical wastewater after treatment	MPC
Be	0,00036	0,00003	0,0002
Al	0,07	0,00600	0,5
Mn	1,3	0,02025	0,1
Ni	66,00	0,08673	0,1
Cu	21,00	0,02249	1,0
Zn	0,076	0,00560	1,0
As	0.88	0,09625	0,05
Se	1.0	0,09723	0,01
Sr	4.8	0,82857	7,0
Mo	2.8	0,00525	0,25
Cd	0.00085	0,00018	0,001
Hg	0.000072	0,00003	0,0005
Pb	0.00013	0,00010	0,03
Th	0.00015	0,00025	-
U	2.4	0,00540	0,7
Cr	0.023	0,02456	0,5
Co	21.0	0,01333	0,1

Considering that the results obtained using model solutions do not always correspond to those obtained with real solutions, it was planned to investigate the sorption of metals present in actual industrial wastewater.

According to the results presented in the table above, the initial composition of metallurgical industry wastewater contains concentrations of metals significantly higher than the MPC, indicating the necessity of wastewater treatment. It can be observed that the concentrations of Ni and Cu ions were reduced several-fold, and even radioactive ions were sorbed to levels below the MPC. [9; pp. 84–86]

It has been demonstrated that wastewater can be purified from metal ions using carbonaceous adsorbents based on activated walnut-shell charcoal and activated almond-shell charcoal, resulting in a reduction of Ni and Cu ion concentrations to levels below the MPC. The obtained results demonstrate that the use of carbon-based adsorbents is one of the effective approaches for removing heavy metal ions from industrial wastewater. In particular, adsorbents prepared from activated walnut-shell and almond-shell charcoal are of considerable interest due to their sorption properties. The use of such adsorbents makes it possible, on the one hand, to reduce environmentally hazardous components in wastewater and, on the other hand, to convert agricultural waste into valuable sorption materials. The complex and variable composition of industrial wastewater is one of the main factors affecting the adsorption process. The concentration of metal ions, pH of the medium, amount of adsorbent, contact time, and presence of other ions can significantly influence the efficiency of the sorption process. Therefore, the sorption characteristics determined using model solutions should be applied to real industrial wastewater with appropriate consideration of the differences in their chemical composition. The experiments conducted with real metallurgical wastewater demonstrated the ability of activated walnut-shell and almond-shell carbon adsorbents to remove metal ions from aqueous media. In particular, the significant reduction in the concentrations of Ni and Cu ions indicates a high sorption activity of the prepared adsorbents toward these metal ions. The obtained results demonstrate the potential of these materials for application in the preliminary or additional treatment stages of industrial wastewater. The production of adsorbents from waste biomass is also important from both economic and environmental perspectives. Instead of disposing of walnut and almond shells as waste, they can be converted into materials with enhanced sorption properties through carbonization and activation. This approach is consistent with the principles of waste recycling, utilization of secondary raw materials, and protection of water resources. The adsorption efficiency of carbon-based adsorbents depends on their surface structure, specific surface area, pore volume, surface functional groups, and the chemical properties of the adsorbate. In the case of industrial wastewater, the adsorption process is additionally affected by the presence of several metal ions and other dissolved substances.

Therefore, the interaction between the adsorbent surface and metal ions should be considered as a complex physicochemical process. The activated carbon obtained from walnut and almond shells contains a developed carbonaceous structure and surface-active sites capable of interacting with metal ions. During the adsorption process, metal ions from the aqueous phase can interact with oxygen-containing functional groups located on the adsorbent surface. Depending on the chemical composition of the wastewater and the properties of the adsorbent, the removal of metal ions may occur through surface adsorption, ion exchange, complex formation, and pore diffusion.

The amount of adsorbent is an important parameter affecting the purification efficiency. Increasing the adsorbent dosage generally increases the number of available active sites and, consequently, increases the percentage removal of metal ions from the aqueous phase. However, excessive amounts of adsorbent may not always result in a proportional increase in adsorption efficiency because a significant part of the active surface may remain unused. Therefore, determining the optimum adsorbent dosage is important for developing an economically efficient purification technology. The contact time between the adsorbent and wastewater also plays an important role in the adsorption process. At the initial stage of adsorption, a relatively rapid decrease in the concentration of metal ions can be observed due to the availability of a large number of free active sites on the adsorbent surface. As the adsorption process proceeds, these active sites become progressively occupied, resulting in a gradual decrease in the adsorption rate until equilibrium is reached. The pH of the aqueous medium is another important factor influencing the adsorption of metal ions. Changes in pH affect both the surface charge of the adsorbent and the chemical form of metal ions in solution. Therefore, the determination of the optimum pH value is necessary to achieve maximum adsorption efficiency. In real industrial wastewater, however, the simultaneous presence of different metal ions may lead to competitive adsorption and affect the removal efficiency of individual components. The obtained results indicate that activated carbon prepared from agricultural waste can effectively interact with metal ions present in industrial wastewater. The decrease in Ni and Cu concentrations after treatment confirms the sorption activity of the prepared carbonaceous materials. Compared with model solutions, real industrial wastewater contains a more complex mixture of dissolved substances; therefore, the results obtained from real wastewater provide more practically relevant information regarding the applicability of the adsorbents. The use of agricultural waste as a raw material for producing adsorbents also provides an additional environmental advantage. Walnut and almond shells are relatively inexpensive and readily available biomass materials. Their

conversion into activated carbon makes it possible to simultaneously address two environmental problems: the utilization of agricultural waste and the purification of contaminated water.

The adsorption capacity of the prepared materials can be further evaluated using adsorption isotherm models, such as the Langmuir and Freundlich models. Kinetic studies based on pseudo-first-order and pseudo-second-order models can also provide information about the mechanism and rate-controlling stages of the adsorption process. Such investigations are important for determining the optimal operating conditions and assessing the possibility of scaling up the proposed purification process. Thus, the obtained experimental results provide a basis for considering activated walnut-shell and almond-shell carbon as promising low-cost adsorbents for the removal of metal ions from industrial wastewater. Further optimization of the activation conditions and adsorption parameters may significantly improve their sorption capacity and expand their practical application in wastewater treatment technologies.

CONCLUSION

In this study, the possibility of reducing metal ions present in metallurgical industry wastewater using a sorption method was investigated. Experiments conducted with both model solutions and real industrial wastewater demonstrated that the efficiency of the adsorption process depends on the initial composition of the wastewater and the amount of adsorbent used. The application of carbon-based adsorbents obtained from activated walnut and almond shells enabled a significant reduction in the concentrations of Ni and Cu ions in wastewater. For certain metal ions, the concentrations were reduced to values close to or below the established maximum permissible concentrations (MPC). These findings indicate that the developed adsorbents are promising materials for the treatment of metallurgical industry wastewater. Overall, the use of activated carbon adsorbents produced from agricultural waste can be considered a relatively simple, resource-efficient, and environmentally sustainable approach to industrial wastewater treatment. Further studies should focus on determining the optimum adsorbent dosage, contact time, pH, adsorption kinetics, and adsorption isotherms, as well as investigating the regeneration and reuse of the adsorbents. Such studies will contribute to improving the practical efficiency and technological applicability of these sorption materials.

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