JOURNAL OF WATER AND LAND DEVELOPMENT
e-ISSN 2083-4535

Polish Academy of Sciences (PAN)
Institute of Technology and Life Sciences (ITP)

JOURNAL OF WATER AND LAND DEVELOPMENT
2021, No. 48 (I–III): 99–104; https://doi.org/10.24425/jwld.2021.136152

Available (PDF): https://www.itp.edu.pl/JWLD; http://journals.pan.pl/jwld

Received 13.01.2020
Reviewed 06.06.2020
Accepted 20.07.2020

Adsorption method of purification of stocks from chromium(III) ions by bentonite clays

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For citation: Soloviy Ch., Malovanyy M., Palamarchuk O., Trach I., Petruk H., Sakalova H., Vasylinych T., Vronska N. 2021. Adsorption method of purification of stocks from chromium(III) ions by bentonite clays. Journal of Water and Land Development. No. 48 (I–III) p. 99–104. DOI 10.24425/jwld.2021.136152.

Abstract

The process of sorption of chromium(III) ions with a stationary sorbent layer of bentonite clays was investigated. The main advantages of using bentonites in water purification technologies are described: powerful geological reserves, cheap process of rock extraction, easy preparation for transportation and use, possibility of using waste sorbents in other technologies that is why there is no need in costly regeneration.

The influence of various factors (process duration, an adsorbent layer) on the degree of wastewater purification from chromium ions, the effect of pumping speed on the dynamic capacity of the sorbent was studied and the effective volume was determined. The adsorption efficacy increases with the increase of the adsorbent layer, what can be explained by the development of the active sorption surface. As the initial concentration of chromium ions increases, the time of appearance of the first traces of the contaminant at the exit of the column increases, as well as the total time to channeling. The results of the studies indicate a higher adsorption capacity of modified bentonite with respect to Cr^{3+} ions compared to its natural formula. The cleaning efficacy of the solution with a concentration of chromium ions of 0.5 $g \cdot dm^{-3}$ is increased by 5% when using 15 g of modified bentonite and 6,5% in the case one uses 20 g compared to the natural form.

Key words: *adsorption, bentonite, chromium ions, environmental safety, natural clay sorbents, wastewater purification*

INTRODUCTION

The problem of environmental protection is extremely important to modern communities although it has been known for a long time. Modern conditions of human life are increasingly demanding and lead to the possible consequences of its activities, not only because the violation of environmental principles forms a circle of irreparable costs in nature, but also because by changing the environment in one direction or another, people directly and indirectly affect the genetic fund, reduce the possibility of a full life.

The objective reality of the modern world indicates that the environmental consequences of human activity are becoming a major factor in the development of a society. It is important to understand and adopt modern value orientations, semantic attitudes, to form a new image of a human being, that would be friendly towards both himself and the natural environment. Without such global psychological restructuring of the human-natural environment, all economic, environmental and scientific-technical measures will be of limited nature and will not be an important basis for overcoming an environmental disaster.

Modern industrial wastewater purification technologies have to meet three requirements which are the following:
1) socio-psychological, aimed at meeting the society's expectations as for environmental situation improvement;
2) economic: price optimization and quality criteria;
3) technological, aimed at reducing material and labour coefficient.

Sorption methods of water purification take the leading place according to the integrated criteria. Some scientists have dedicated their studies over the last few years to heavy metal adsorption with the help of dewatered iron-containing waste sludge [YILDIZ *et al*. 2018]. Considerate attention is also being paid to studying on the use of ferruginous sludge from water treatment plants [WOŁOWIEC *et al*. 2017].

It should be noted that heavy metals form a group of the most dangerous pollutants in the environment. In surface natural reservoirs (seas, lakes, rivers, reservoirs) with industrial wastewater, a considerable amount of heavy metal ions enters, which has become a significant obstacle in the life of macrobiota. The wastewater of the electroplating, paint and leather industries contain a large number of soluble and insoluble heavy metal compounds that have an unpleasant odour, dark colour, foam and are toxic. The content of chromium(III) ions, for example, in sewage production of leather can reach 3000 or more $mg \cdot dm^{-3}$ [PETRUS, WARCHOŁ 2005]. Given the toxicity of this metal, wastewater containing its compounds is subject to mandatory purification prior to their discharge into natural reservoirs.

The compounds of tri- and hexavalent chromium exhibit the opposite physiological effect. If micro amounts of Cr(III) compounds play an important role in mammalian metabolism, then Cr(VI) compounds have toxic effects on biological systems and carcinogenic effects on humans. Compared to Cr(III) compounds, chromium(VI) compounds are soluble in a wide pH range, have greater mobility and are therefore more dangerous to living organisms. In addition, the harmful effects of hexavalent chromium are exacerbated due to the fact that the negatively charged ions $HCrO_4^-$, CrO_4^{2-}, $Cr_2O_7^{2-}$ limited undergo sorption by clays, which increases their mobility and enhances the hazardous effect [SAKALOVA *et al*. 2017]. Ranges from a few tenths of a microgram in a dm^3 to a few micrograms in a dm^3 in polluted ponds it reaches several tens and hundreds of micrograms in a dm^3. The average concentration in seawater is 0.05 $mg \cdot dm^{-3}$, and in groundwater it is usually within $n \cdot 10$–$n \cdot 10^2$ $mg \cdot dm^{-3}$ [MALOVANYY *et al*. 2019]. The chromium content of reservoirs for drinking purposes shall not exceed the maximum permissible concentration (MPC) for Cr(VI) of 0.05 $mg \cdot dm^{-3}$. The MPC in the water of the reservoir used for fishery purposes for Cr(VI) is 0.001 $mg \cdot dm^{-3}$.

One of the promising ways of manifestation of greening is the study of sorption separation as a method of extraction of radioactive, toxic and valuable impurities from aqueous solutions. Ion-exchange methods have particular advantages in cases where the initial concentration of unwanted impurities is relatively small and the degree of their extraction should be high.

In terms of prospects for the application of innovative methods of treatment of chromium-containing wastewater in the leather and fur industry, in comparison with existing (reagent electrochemical, membrane, ion exchange methods) the most effective today are sorption methods. The use of active sorbents allows to thoroughly treat effluents that contain even small concentrations of these metals, and it is known that the content of Cr_2O_3 in wastewater after tanning and before duplication is relatively small, and ranges from 0.5–2.2 $mg \cdot dm^{-3}$. Sorbents of natural origin are increasingly used for water purification: clay rocks, apatites, zeolites. The use of such sorbents is due to their relatively high sorption capacity, cation exchange properties of some of them, relatively low cost and availability.

Analysis of recent publications (WARCHOŁ *et al*. [2006]; KOŁODYŃSKA *et al*. [2017]) indicates the feasibility of using adsorption methods for the purification of wastewater from heavy metal ions using natural dispersed minerals as adsorbents [KONEFAŁ *et al*. 2015], as modification objects [ILANGO, NATRAYASAMY 2018; REPO *et al*. 2017], or carriers for mycelium fungi *Aspergillus* immobilization [SATHVIKA *et al*. 2015]. Modified natural sorbents have improved properties concerning heavy metals [REPO *et al*. 2013], and immobilized on sorbents fungi *Aspergillus* are used for purifying effluents from hexavalent chromium. Quite often, silica gels [REPO *et al*. 2011], clay materials [ANNAN *et al*. 2018; QIN *et al*. 2016], humic complexes [MENG *et al*. 2017] are used to purify wastewater from heavy metal ions. Purification of aqueous solutions with the help of dispersed sorbents is in accordance with the principle of environmental life of modern man, that is, indicates compliance with the requirements of environmentally friendly and energy-saving production [PALAMARCHUK 2011]. Powerful geological reserves, cheap rock extraction, easy preparation for transportation and use, the ability to use waste sorbents in other technologies are the main advantages of using natural minerals. One of the types of promising natural sorbents, which have been used as an object of research, is bentonite – a natural clay sorbent with a layered structure. The main mineral that provides the adsorption properties of bentonite is montmorillonite.

The main advantages of using bentonites for the treatment of chromium-containing effluents include:
– natural bentonites are widespread in Ukraine;
– natural sorbents are an affordable, inexpensive materials;
– adsorption technologies using bentonites provide a high degree of wastewater treatment from heavy metal ions;
– spent natural adsorbent can be disposed within the technologies of leather and fur production.
– bentonite saturated with chromium ions can be reused for wastewater treatment of preparatory processes and spent nickel plating fluids.

MATERIALS AND METHODS

The purpose of the work is to study wastewater treatment of chromium (III) ions by adsorption on natural sorbents. Studies of the adsorption process, as a manifestation of environmentally oriented activity, have been carried out in an adsorption column with a fixed layer of adsorbent. The proposed technical solutions make it possible to significantly simplify and reduce the cost of sewage treatment in various industries, including galvanic and leather industries.

Bentonite (type 2:1) from the Cherkasy deposit of bentonite and Paligorskite clays was used for the research. The preparation process for bentonite consists of several steps. The raw material was crushed and dried in a drying chamber at a temperature of 120°C for 45 min. Then the clay was ground and separated into appropriate fractions. Water was purified on a laboratory adsorption column having a diameter of 35 mm and a height of 300 mm using bentonite clay. The prepared clay was poured into a column. The total weight of the sorbent in the adsorption column was 15–25 g. The volume of the sorbent in the column was 21.6 cm^3 and 29 cm^3, respectively, by the weight of the sorbent 15 and 20 g (usually to make it more clear it has been used OK unit "column volume" – standard volume of column for the same weight of sorbent). In fact, this is a dimensionless quantity, which is denoted by the indicator V_{eff}. The solutions containing the content of Cr^{3+} ions of the given concentration were passed through the column. During the sorption samples were taken every 10 cm^3, the concentration of Cr^{3+} ions in the solution was determined by the titrimetric method. To establish the range of mode parameters which require a detailed study of adsorption processes, a series of preliminary experiments was performed which made it possible to draw the following preliminary conclusions:

- fluctuations in temperature from +10 to +30°C have no significant effect on the degree of adsorption of Cr^{3+} bentonite ions;
- the optimal average pumping rate of the model solution through the adsorption column of the given height is 0.3–0.5 $cm^3 \cdot min^{-1}$, at higher speed the amount of adsorbed Cr^{3+} is reduced and the lower speed increases the probability of a strong thickening of the pulp which complicates the processes of further filtration;
- the interval of investigated concentrations (0.5–2.0 $g \cdot dm^{-3}$) of chromium ions was chosen based on practical considerations in accordance with the possible content of Cr^{3+} cation by real effluents;
- it is preliminary established that the complete saturation of bentonite clay with Cr^{3+} ions is achieved at a layer of adsorbent 15–20 g, for 1–3 days depending on the concentration of the model solution.

To determine the dependence of the sorption efficiency on the concentration of chromium(III) ions, model solutions with an initial concentration of pollutant from 0.5 to 2 $g \cdot dm^{-3}$ were passed through an adsorption column at 15 g adsorbent layers; 20 g; the temperature of the model solution was 20°C. The solutions were analysed through each column volume, the average passage time was 3–4 $min \cdot cm^{-3}$, the control points were determined every 40 cm^3 of the solution.

RESULTS AND DISCUSSION

As shown by the saturation curves of bentonite (Fig. 1) by chromium ions, presented in the form of the concentration of heavy metal ion at the outlet of the column (C_{eff}) on the volume of pumped model solutions (V_{eff}), the adsorbent consumption has a significant influence on the course of the saturation process. With the same concentrations of chromium ions, the volume of the solution and the adsorption time before the full saturation of the adsorbent are much higher. The highest effective volume when pumping a model solution through a 15 g sorbent layer is 30.3, and when passing a solution through 20 g sorbent it is 41.4 cm^3. These values in both cases are observed for model solutions with the highest concentration of heavy metal ion and this suggests that the use of a fixed-bed adsorbent adsorption method to remove low concentrations of pollutants (less than 0.5 $g \cdot dm^{-3}$) is possible at lower bentonite costs. Also, with an increase in the initial concentration of chromium ions within the experimental values, the time of appearance of the first trace of the pollutant at the exit of the column increases, and the time to slip in all cases quickly occurs at the concentration of the pollutant at the

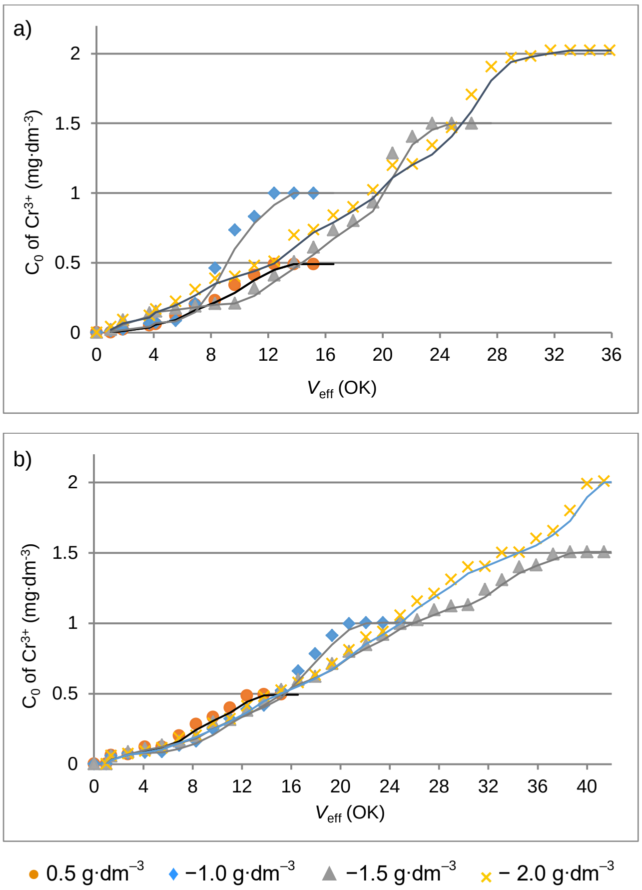

Fig. 1. Saturation curves of bentonite with a model solution with different concentration of Cr^{3+} (C_0): a) bentonite weight 15 g, b) bentonite weight 20 g; source: own study

exit from the column 70% of the initial one. The exceptions are the dependence on the concentration of chromium ions (C_{Cr}) of 0.5 g·dm^{-3} – in this case, the increase in the concentration at the outlet of the column is linear.

According to the results of the experiment, the maximum absorption of chromium(III) ions is 70.2–83.2% for the consumption of sorbent weighing 20 g and 63.5–82.2% in the case of mass of sorbent 15 g (Tab. 1). The adsorption efficiency increases with the increase of the adsorbent layer, which can be explained by the development of the active sorption surface.

Table 1. Indicators of sorption efficiency at different initial concentrations of chromium(III) ions in solution

Time (min)	Initial concentration C_0 of Cr^{3+} (g·dm^{-3})	The first traces Cr^{3+} in the purified solution (OK)	Effective volume V_{eff} (OK)	Dynamically exchange capacity (T)	α (%)	
					a_{max}	$\bar{a}$
Weight of bentonite 15 g						
1080	0.5	1.39	11.03	0.001	95.6	82.2
1230	1.0	1.39	12.41	0.002	97.5	81.0
2480	1.5	0.92	23.45	0.002	98.6	74.0
3205	2.0	0.92	31.72	0.003	98.9	63.5
Weight of bentonite 20 g						
1380	0.5	1.03	13.79	0.0008	95.9	83.2
2520	1.0	0.69	22.07	0.0010	97.8	80.2
3720	1.5	0.69	38.06	0.0015	98.7	78.3
4100	2.0	0.69	41.38	0.0020	98.9	70.2

Source: own study.

The small values of the dynamic exchange capacity are due to the fact that the first traces of the pollutant are already defined by 1–2 OK, and this value is much lower than in the case of pumping model solutions with ions of other pollutants [PETRUSHKA *et al.* 2014], although the degree of purification of the first volumes is high – 95.6–99% for solutions with different concentrations of Cr^{3+}. The first traces of chromium ions appear earlier when using 20 g of bentonite and the values of the dynamic exchange capacity are slightly lower when using more adsorbent. However, the gains in the larger dynamic exchange capacity are small compared to the difference between volumetric pumping rates. So the difference between the volumes of pumped runoff before the "breakthrough" is much higher when using 20 g of bentonite, and in the case of concentrations greater than 0.5 g·dm^{-3} this difference is 10 and above OK. Thus, the study of adsorption extraction of chromium(III) ions by bentonite clays confirms the ability of modern humans to navigate environmentally sound behaviour and demonstrate an active attitude towards the development of new environmental standards.

With the use of modified bentonite [SAKALOVA *et al.* 2019; VASYLECHKO *et al.* 2003], the saturation curves indicate similar dependences: the adsorption capacity of bentonite is higher in case of lower concentrations of the stock solution, and if one uses 20 g of adsorbent, the purification efficacy is higher. The time intervals of the control measurements are very close what can be proved, as well by similar values of the effective volume indicator, but the first traces of pollutant at the exit of the column appear later (Tab. 2).

Table 2. Efficacy indicators of sorption of chromium(III) ions with the concentration of 0.5 g·dm^{-3} on modified bentonite

No.	Time (min)	The first traces (Cr^{3+}) in the purified solution (OK)	Effective volume V_{eff} (OK)	Dynamical exchange capacity (T)	α (%)	
					a_{max}	$\bar{a}$
Weight of bentonite 15 g						
1	1000	1.86	11.00	0.001	92.8	87.2
Weight of bentonite 20 g						
2	1220	1.30	12.47	0.008	96.9	89.7

Source: own study.

The results of the studies indicate a higher adsorption capacity of modified bentonite with respect to chromium ions compared to its natural formula. The cleaning efficacy of the solution with a concentration of chromium ions of 0.5 g·dm^{-3} is increased by 5% when using 15 g of modified bentonite and 6.5% in the case one uses 20 g compared to the natural form. It is important to mention that the modification of bentonite can be carried out by pre-soaking it in the spent acid solution [SABADASH *et al.* 2017], for example, in a solution after nickel plating that has undergone mechanical cleaning.

The proposed wastewater treatment method from chromium(III) ions may find implementations for wastewater treatment in various industries. We have considered the application of the method in the leather industry. In our opinion, one of the ways of using the method can be carried out in the following scheme (Fig. 2).

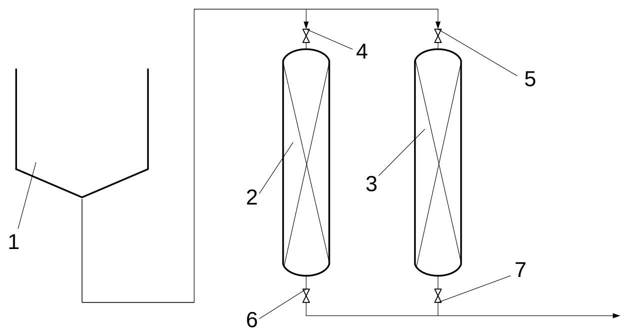

Fig. 2. Scheme of wastewater treatment from chromium(III) ions; source: own study

The water with chromium(III) ions from collector *1* are fed to the adsorption columns *2* and *3*. The columns work by shift: one of the columns operates in adsorption mode, the other in adsorbent overload mode. Purified effluents emanating from the adsorption column are used as industrial water in the leather production process. The column in adsorption mode works to saturate the adsorbent with chromium ions, which leads to a breakthrough at the exit of the column. After locking of the breakthrough, the columns are switched with the help of valves *4*, *5*, *6* and *7*. The sewage for purification is fed to another column, and the column, which worked in the adsorption mode, is unloading the spent adsorbent and loading a fresh portion of bentonite. The spent adsorbent (saturated with chromium(III) ions bentonite) can be used in leather production for tanning and filling of natural leather [PALAMAR *et al.*

2015]. We also conduct further research on the use of waste sorbents as components of the aniline coating and pigment mixture in the final treatment of natural leather.

CONCLUSIONS

The effectiveness of the use of natural mineral sorbents, in particular bentonite clays, for wastewater treatment is confirmed by their advantages over other sorbents, namely: they benefit in accessibility, cost, regeneration and reusability. Studies have confirmed the importance of modern human environmental activity, which is implemented in the system of target ecological installations, value ecological relationships and environmental orientations, which determines the prospect of using bentonite clays for wastewater treatment of chromium(III) ions by adsorption.

According to the results of the experiment, the efficiency of adsorption increases with increasing layer of adsorbent. Thus, the maximum absorption of chromium(III) ions occurs at a layer of sorbent 20 g and low concentrations of pollutant and is 83.2%.

The degree of wastewater purification during adsorption with a fixed sorbent layer at different concentrations of chromium(III) ions and quantities of the sorbent was determined. The bentonite consumption was found to be 20–22 $g \cdot dm^{-3}$ of contaminated water to purify the solution containing Cr^{3+} ions at a concentration of 1–2 $g \cdot dm^{-3}$.

The basic technological scheme of sewage treatment of leather production from chromium(III) ions is offered. Processed adsorbent saturated with chromium ions can be used in leather production for tanning, filling and finishing of natural leather.

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