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THE USE OF LEAVES OF DIFFERENT TREE SPECIES AS A SORPTION MATERIAL FOR EXTRACTION OF HEAVY METAL IONS FROM AQUEOUS MEDIA

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Received on 08-06-2016

Accepted on 29-06-2016

Abstract

In a review article summarizes the literature data on the use of tree leaves as sorption materials for removal of heavy metal ions from aqueous media. It is shown that the sorption characteristics of the leaves of trees and the degree of removal of metal ions depends both on the conditions of adsorption (initial concentration of metal ion, temperature, pH, duration of contact), and from species trees. It is shown that the majority of publications devoted to removal from aqueous media toxic ions Cd(II), Cr(VI), Cu(II) and Pb(II). The greatest sorption performance removal of metal ions and the largest number of publications in the literature devoted to sorption using the leaves of the Neem tree (*Azadirachta indica*).

Keywords: Leaves, Heavy metals, Sorption.

Introduction

Currently, the contamination of environment by various pollutants, including metal ions, represents a great danger to the biosphere. In addition to direct toxic action on living and plant organisms ions metals tend to accumulate in the food chain, which increases their risk to humans. Once in the water, they stay for a long time in the most dangerous ionic form, and even passing in the bound state (colloidal form, bottom deposits or other soluble compounds), continue to pose a potential threat to biological objects.

There are many different chemical, physical and biological methods for the recovery of metal ions from aqueous media. As the review of the world literature, is currently one of the most effective methods for removing pollutants from aqueous media is sorption. The special value of this method is that as sorption materials can be used waste products of industrial production and processing of agricultural products, as well as natural plant biomass.

Of particular interest is the use as reagents for the purification of natural and waste water renewable woody biomass.

In world literature there are a large number of publications devoted to the removal of pollutants from aqueous media waste of wood processing, usually sawdust [1]. At the same time, very promising sorption materials is the foliage of the trees.

In the present work summarizes literature data on removal of metal ions from aqueous media using tree leaves.

A simple leaf consists of a single leaf and a single petiole. Depending on the types of trees, their leaves can be simple or complex, have a different shape, venation, but the internal structure all the same. The leaf of the tree consists of an outer cuticle, stomata and underlying tissue. The upper skin (epidermis) – integumentary tissue on the facing side of the sheet, often covered with hairs, cuticles. The stoma – slit surrounded by cells through which air is supplied to the internal cells of the leaf through them gaseous substances, including water vapor out of the leaf outward. To the underlying tissues of the leaf are: columnar tissue, the cells of which are cylindrical, densely adjoin to each other and are located on the upper side of the sheet (facing the light), spongy tissue, the cells of which have a rounded shape, are arranged loosely between them and formed large intercellular spaces filled with air, conductive fabric, riddled with veins. Veins is spending bunches, as they formed the conducting tissue – the phloem and wood.

In addition to the phloem in the composition of the conductive beam enters and wood. Through the vessels of the leaf, as in the root, moving water with dissolved mineral substances. Then from the roots through the vessels of wood these substances enter in aerial organs, including the cells of the sheet.

In the numerous veins include fiber. It's a long cell with pointed ends and thick lignified membranes. Major leaf veins are often surrounded by mechanical tissue, which consists entirely of thick-walled cells called fibers.

Lower the skin integumentary tissue on the lower side of the leaf, usually bears stomata [2].

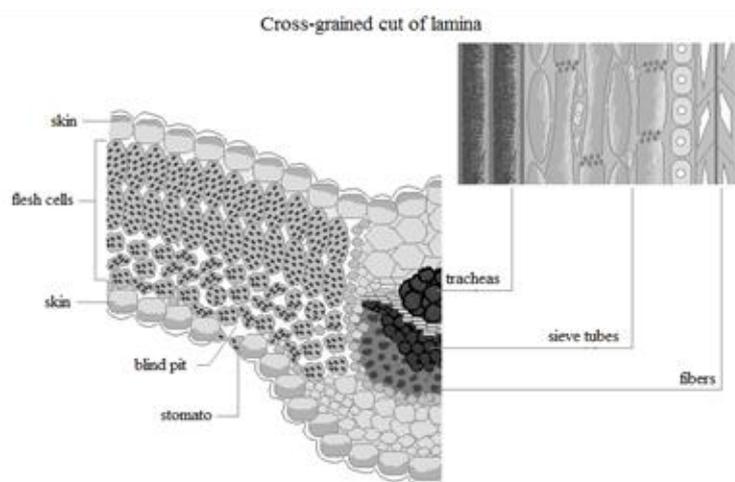


Figure 1. Cross section of lamina.

Chemical and elemental composition of the leaves depends on the species, some of the elements found in the composition of the leaves can also depend on the location of growth, as is often the bioindicators for environmental monitoring. Table 1 provides information about the composition of the leaves of *Betula pendula* and *Pópulus trémula* [3], the most widely distributed in Central Russia.

As can be seen from the data presented in table 1, the sheets named trees have different content components, especially different contents of lignin and pentosan for the same amount of cellulose. The presence of leaf lignin and cellulose, which include a large number of different functional groups, as well as the developed surface, gives a material called high sorption performance.

Table 1. Chemical composition of leaves of birch and aspen.

The component parts of the leaves	<i>Betula pendula</i> Leaves, % of dry sample	<i>Pópulus trémula</i> Leaves, % of dry sample
Cellulose	25,2	25
Lignin	34,2	29,5
The pentosans	17,9	11,2
Polihronova acid	12,2	11,55
Substances soluble in hot water	32,5	29
Substance, soluble in sulphuric ether	8,42	7,96
Substances that are soluble in a mixture spiropentane	14,7	17,5
Methoxyl group (-OCH ₃)	2,3	2,55
Ash contents	4,22	5,65
Easy hydrolysable materials (PB)	17,6	14,9
Methoxyl group in xylogen	2,77	4,02

To date, studied the sorption characteristics of the foliage of different tree species in relation to ions of metals. A list of references, depending on the metal cation and tree species are shown in table 2.

Table 2. Sorption properties of different sorts wood leaves in relation to metals.

Heavy metal	Sorts wood	References
1	2	3
Cd ⁺²	Neem (<i>Azadirachta indica</i>)	[4], [20], [21]
	Guava (<i>Psidium guajava</i>)	[5], [6], [7]
	Jambolan (<i>Syzygium cumini</i>)	[8], [9]
	Ulmus (<i>Ulmus</i>)	[10]
	Myrtle (<i>Myrtus</i>)	[11]
	Loquat (<i>Eriobotrya japonica</i>)	[12]
	<i>Mangifera indica</i> (<i>Mangifera indica</i>)	[13]
	Hornbeam (<i>Carpinus betulus</i>)	[14]

	Cherry (<i>Prunus avium</i>)	[15]
	Wiliwili (<i>Erythrina sandwicensis</i>)	[16]
	Bengal almond (<i>Terminalia catappa</i>)	[17]
	Moringa oleifera (<i>Moringa oleifera</i>)	[18], [19]
	Olive (<i>Olea europaea</i>)	[22]
	Mistletoe (<i>Viscum</i>)	[23]
	Teak (<i>Tectona grandis</i>)	[24]
	Sacred fig (<i>Ficus religiosa</i>)	[25]
	Nipa palm (<i>Nypa fruticans</i>)	[26]
1	2	3
Cu ²⁺	Camphorwood (<i>Cinnamomum camphora</i>)	[27]
	Vachellia nilotica (<i>Acacia nilotica</i>)	[28]
	Neem (<i>Azadirachta indica</i>)	[29], [30]
	Doum palm (<i>Hyphaene thebaica</i>)	[31]
	Mangifera indica (<i>Mangifera indica</i>)	[19]
	Moringa oleifera (<i>Moringa oleifera</i>)	[32]
	Loquat (<i>Eriobotrya japonica</i>)	[33]
	Olive (<i>Olea europaea</i>)	[22]
	Teak (<i>Tectona grandis</i>)	[34],[35]
	Indian coral tree (<i>Erythrina variegata</i>)	[36]
Pb ²⁺	Bael (<i>Aegle marmelos</i>)	[37]
	Jambolan (<i>Syzygium cumini</i>)	[38]
	Camphorwood (<i>Cinnamomum camphora</i>)	[39]
	Catalpa (<i>Catalpa speciosa</i>)	[40]
	Sacred fig (<i>Ficus religiosa</i>)	[41]
	Loquat (<i>Eriobotrya japonica</i>)	[33]
	Neem (<i>Azadirachta indica</i>)	[21],[42],[43]
	Olive (<i>Olea europaea</i>)	[22]
	European mistletoe (<i>Viscum album</i>)	[44]
	Mistletoe (<i>Viscum</i>)	[23]
	Robinia (<i>Robinia pseudoacacia</i>)	[45], [46]
	Chinese parasol tree (<i>Firmiana simplex</i>)	[47]
	Rubber tree (<i>Hevea brasiliensis</i>)	[48]
	Cashew tree (<i>Anacardium occidentale</i>)	[49]
	Papaya tree (<i>Carica papaya</i>)	[49]
	Mahua (<i>Madhuca longifolia</i>)	[50]
	Asopalav tree (<i>Polyalthia longifolia</i>)	[50]
	Sesame (<i>Sesamum indicum</i>)	[51]
	Mangifera indica (<i>Mangifera indica</i>)	[13]
Zn ²⁺	Jambolan (<i>Syzygium cumini</i>)	[52]
	Teak (<i>Tectona grandis</i>)	[53]
	Neem (<i>Azadirachta indica</i>)	[54],[55]
	Indian coral tree (<i>Erythrina variegata</i>)	[36]
Cr ⁶⁺	Neem (<i>Azadirachta indica</i>)	[56],[60], [61],[63]
	Gum arabic tree (<i>Acacia nilotica</i>)	[57]
	Nipa palm (<i>Nypa fruticans</i>)	[26]
	Castor oil plant (<i>Ricinus communis</i>)	[58]
	Mangifera indica (<i>Mangifera indica</i>)	[59], [6]
	Gum trees (<i>Eucalyptus</i>)	[61]
	Mangroves (<i>Rhizophora mangle</i>)	[62]
	Euclea schimperi (<i>Euclea schimperi</i>)	[64]
Indian coral tree (<i>Erythrina variegata</i>)	[65]	
1	2	3
Co ²⁺	Gum arabic tree (<i>Acacia nilotica</i>)	[66]

	Teak (<i>Tectona grandis</i>)	[67]
Ni ²⁺	<i>Mangifera indica</i> (<i>Mangifera indica</i>)	[13]
	<i>Moringa oleifera</i> (<i>Moringa oleifera</i>)	[19]
	Teak (<i>Tectona grandis</i>)	[67]
As ³⁺	Ash (<i>Fraxinus</i>)	[68]
	<i>Moringa oleifera</i> (<i>Moringa oleifera</i>)	[69]
Hg ²⁺	Ash (<i>Fraxinus</i>)	[70]
	Castor oil plant (<i>Ricinus communis</i>)	[71]
Ce ³⁺	Oriental plane (<i>Platanus orientalis</i>)	[72]
La ³⁺	Oriental plane (<i>Platanus orientalis</i>)	[72]
Ag ²⁺	Centuryplant (<i>Agave americana</i>)	[73]
	Judas tree (<i>Cercis siliquastrum</i>)	[74]
Tl ³⁺	Ulmus (<i>Ulmus</i>)	[75]
	Gum trees (<i>Eucalyptus</i>)	[76]
Au ³⁺	European nettle tree (<i>Celtis australis</i>)	[77]
	Paulownia (<i>Paulownia</i>)	[77]
	Sweet chestnut (<i>Castanea sativa</i>)	[77]

As can be seen from the above data, the majority of publications devoted to the extraction of heavy metal ions from aqueous media by the leaves of trees of various species. The degree of removal of each metal ion from the water depends primarily on the nature of the metal cation and the type of foliage and conditions of the sorption process (initial concentration of metal ion, pH, temperature, dosage of the sorption material, the sorption time).

The removal of ions Cd(II)

The greatest number of publications devoted to the removal of ions Cd²⁺. In particular, studies found that the degree of removal of ions Cd(II) by leaves of the tree *Azadirachta indica* ranges from 86 to 95 %, the smaller the degree of extraction of named ions is observed when the leaves of *Psidium guajava* - 60-63 % [4-6]. The greatest degree of removal of ions Cd(II) (98 %) is observed in the case of using the leaves of the tree *Syzygium cumini* [8]. Conducted experiments have shown that depending on the wood species and the experimental conditions, the maximum sorption capacity of the leaves for ions Cd(II) is 15-48 mg/g [5, 8, 9, 10, 13], and depends on the pH of the aquatic environment. Table 3 shows the values of maximum sorption capacity (Q_{max}) of some of biosorbents with respect to ions of Cd(II).

Table 3. Maximum sorption capacity of various biosorbents with respect to ions of Cd(II).

Biosorbent	Q_{max} mg/g
Leaves <i>Ulmus</i>	6.94
Leaves of guava	31,15
Leaves <i>Azadirachta indica</i>	21,45
Leaves <i>Syzygium cumini</i>	34,54

Crusts of pomelo	21.83
Olive pits	6.97
Nut plug	42.71
Rice husk	8.58
Halm wheat	11.6

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the sorption capacity of the leaves of the trees for ions Cd(II) above such, for some specific waste processing of agricultural raw materials, behind only the husk of peanuts.

Determined the thermodynamic parameters of interaction between the foliage of trees of various species of ions with Cd(II) [4, 12, 25] (table. 4).

Table 4. Thermodynamic parameters of sorption of ions Cd(II) by leaves of the trees when temperatures 293-313 °K

Sorts wood	ΔH^0 , kJ/mol	ΔS^0 , kJ/mol·K
Jambolan (<i>Syzygium cumini</i>)	3,7	16,87
Neem (<i>Azadirachta indica</i>)	73,7	0,24
Loquat (<i>Eriobotrya japonica</i>)	29,7	125,44
Sacred fig (<i>Ficus religiosa</i>)	8,31	38,22

The data presented in table 4 are ambiguous, because the performance of entropy and enthalpy are defined at different narrow temperature intervals. Varying the experimental conditions, including the initial concentration of the ions in the water.

To increase the sorption capacity of the leaf litter using a modification of the various reagents. In particular, it is shown that the processing of the leaves of the tree *Myrtus* KMnO₄ solution led to increase the degree of purification from 78 % to 98 % at a neutral pH value and contact time 90 minutes [11]. In General, the sorption process is carried out in a weakly acidic medium at pH = 4,5-6,0.

The removal of ions Cu(II)

Extracting ions Cu(II), according to the literature, occurs quite effectively and is 34-90 %. The pH, which was determined by the sorption capacity is in the range from pH = 4-7. The results of the processing of the leaves of *Azadirachta indica* NaOH: when processing the degree of purification increases from 58 to 77,4 % and depends on the contact time between the sorption material with the environment [29, 30]. The greatest degree of cleaning shows the leaves of *Eriobotrya japonica* treated with NaOH – 98 %, despite the fact that the original foliage gives only 68 %

degree of purification. The maximum sorption capacity of the leaves of certain tree species for ions Cu(II) and thermodynamic parameters of adsorption are given in table 5.

Table 5. Indicators of sorption of copper ions (II) the foliage of trees of different species.

Sorts wood	Qmax mg/g	Degree of deionization, %	ΔH_0 , kJ/mol	ΔS_0 , kJ/mol·K	References
Azadirachta indica	21,0-46,0	34-58	15,43	0,116	[29,30]
Azadirachta indica, modified NaOH	50,6-78,0	77-59	-	-	[29,30]
Tectona grandis	95,4- 116,78	72	62,42	0,219	[34,35]
Acácia nilótica	82,63	78	12,345	39,46	[28]
Mangifēra īndica	100	90	-	-	[19]

As seen from table 5, some of the leaves are indicators of the degree of purification is represented by intervals of values. This fact is due to different initial concentrations of ions Cu² in water and the temperature of the process. In addition, the influence on the degree of purification and the maximum sorption capacity is having the time of contact with the sorption material with ions of Cu(II): with increasing time, both indices increase [28-30, 33-35]. In some cases, when the contact time exceeds 150 minutes, there is a decrease in the sorption capacity of the material, due to desorption [29].

The removal of ions Pb(II)

The literature presents a lot of data on removal of lead ions by the foliage of trees. It is shown that the optimal pH for obtaining the maximum extraction of ions Pb(II) – pH = 4-7, the temperature of the process – 20-30 0C, contact time of the sorption material with a sorbate is 60-120 minutes.

The optimum sorption conditions for different types of foliage are different. So, when extracting ions Pb(II) leaves of *Cinnamomum camphora* at a temperature of 30 0C maximum sorption capacity is 73 mg/g [39], and the leaves *Viscum* under the same conditions maximally absorb 769 mg/g [23].

As considerably high sorption capacity to ions of Pb(II) show the leaves of *Azadirachta indica* – 300 mg/g at a temperature of 27 °C [43].

The lowest sorption capacity has foliage of *Catalpa speciosa* is 14.7 mg/g [40]. However, the foliage of different tree species show a relatively high degree of purification: *Azadirachta indica* – 93 % [42], *Hevea brasiliensis* at 95.3 % [48], *Anacardium occidentale* - 92 % [49].

The maximum sorption capacity of the leaves of various species in relation to ions of Pb(II) are shown in table 6.

Table 6: Values of maximum sorption capacity of the leaves of different tree species for ions Pb(II).

Sorts wood	Qmax mg/g	References
<i>Syzygium cumini</i>	32,47	[38]
<i>Cinnamomum camphora</i>	73	[39]
<i>Catalpa speciosa</i>	14,7	[40]
<i>Azadirachta indica</i>	91,34	[21],[42],[43]
<i>Víscum</i>	769	[23]
<i>Firmiána simplex</i>	136,7	[47]
<i>Anacardium occidentale</i>	91,52	[49]
<i>Mangifēra indica</i>	45	[13]

To enhance the sorption of lead ions were processed leaves of different reagents. Thus, in particular, as a reagent for the modification of the leaves of *Hevea brasiliensis* is used an aqueous solution of citric acid [48].

The removal of ions Zn (II)

The extraction of ions Zn (II) from polluted water using tree leaves studied a little, unlike other ions of heavy metals. For example, it is shown that the maximum sorption capacity of the leaves of *Tectona grandis* – 16,42 mg/g, and *Azadirachta indica* – 147,08 mg/g [to 53.54]. The adsorption capacity of the leaves of *Erythrina variegata* with increasing pH from 1 to 6 varies with 38,25 mg/g to 44,77 mg/g. It is shown that with increasing temperature the sorption capacity of the leaves of *Erythrina variegata* decreases, and the sorption curves are described by equations of Langmuir, Freundlich and Temkin [36].

The removal of ions Cr (VI)

The removal of ions Cr(VI) is different from the previous removal of ions (Cd(II), Cu(II), Pb(II)), mainly because in most data the process is carried out in acidic medium at pH = 2-3. In this case, the ions Cr(VI) ions to Cr(III). I.e., occur simultaneously two processes – recovery of ions Cr(VI) and sorption of ions Cr(III). However, there are also data of experiments processed at pH closer to neutral. It is shown that the sorption of chromium ions leaves *Azadirachta indica* occurs at pH = 4.5 to 7.5 and the degree of purification reaches of 99.7 % [60]. The maximum

A. A. Alekseeva* et al. *International Journal Of Pharmacy & Technology*
absorption capacity is 125,83 mg/g [56]. The leaves of *Ricinus communis* as sorbed ions Cr (VI) at pH =7, but the degree of purification when it reaches 59 %. Significantly high degree of purification show leaves *Mangifera indica*, *Eucalyptus*, *Euclea schimperi* – 98-99 %, at pH = 2 and sorption time not exceeding 120 min [61, 64]. For removal of chromium ions (VI) and other heavy metal ions, is used treatment of leaves with different reagents. Shown in particular that the leaves of *Azadirachta indica* after treatment with HCl solution, and the leaves *Acacia nilotica* after modifying solution of H₃PO₄, increase several times the maximum sorption capacity of chromium ions (pH = 2, temperature - 30 °C [57, 63]).

Sorption of ions of precious, rare and other metals

In the literature provides some information on extraction from water bodies of ions of precious metals. For example, in [77] shows the data for the recovery of Au ions in aqueous solutions 16 types of leaves of different trees. It is determined that the degree of removal of gold ions is 80.1-to 98.4% in the interval of pH = 1-4. In this case, the absorbed ions Au(III) in the pores of the sorbent to form compounds that are proposed to extract heat treatment at t = 1200 °C. the study of the sorption of ions of Ag(III) leaf litter little studied, however, in the literature there is evidence that the optimum pH value at which the leaves of *Agave americana* and *Siliquastrum orbicularis* effectively absorb ions Ag(II) is pH = 4.5-6 [74].

The removal of ions As(III) with an efficiency of 73 % can be produced by the leaves of the *Moringa oleifera* or *Fraxinus*, while the foliage of the latter has a maximum sorption capacity of 99.97 mg/g [68, 69]. Thermodynamic sorption characteristics of arsenic leaves of *Moringa oleifera* are $\Delta H^0 = 608,42$ kJ/mol, $\Delta S^0 = 20,35$ kJ/mol·K [69].

Investigated the extraction of ions of cerium and lanthanum leaves of *Platanus orientalis*. It is determined that at t = 50 °C and pH = 6,8 degree of purification from ions of Ce(III) and La(III) is 98,36 % and 86,03 %, respectively [72].

Studied the sorption of ions of thallium (III) leaves of *Ulmus minor* and *Eucalyptus* in a neutral environment. Found that the maximum sorption capacity for ions of Tl(III) leaves of *Ulmus minor* is 54.6 mg/g, leaves *Eucalyptus* – 80,65 mg/g [75, 76]. The degree of purification from ions of thallium in both cases over 80 %.

Conclusions

Summarizes literature data on the use of tree leaves as sorption materials for the extraction of metal ions from aqueous media. It is shown that the sorption characteristics of the leaves of various trees in relation to ions of metals, as well as other sorbents depend on environmental conditions. Basically, the effective sorption takes place in acidic and neutral medium, and rarely in acid. The temperature effect is ambiguous, unlike the pH of the medium. It is

shown that the increase in the sorption capacity of the leaves of trees are possible when processing the last of the various reagents. Studies determined that sorption isotherms of metal ions are described mainly by the equations of Freundlich and Langmuir, at least – Dubinin-Radushkevich and Temkin.

Acknowledgements

The work is performed according to the Russian Government Program of Competitive Growth of Kazan Federal University.

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