Use of chestnut sheel (Castánea) as adsorption material for removing pollutants from natural and sewage waters: a review

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Abstract. The literature data on the use of grinded chestnut shells as a sorption material for removing heavy metal ions, dyes and some organic compounds from aqueous media are summarized. It is shown that a large number of publications are devoted to the study of the adsorption of ions of heavy metals with the shell of chestnut fruits. Information on the chemical composition of the chestnut shell is given. It was shown that the chestnut fruit contains cellulose, lignin and sugars in the shell most of all. It is indicated that the adsorption of many metal ions is carried out by functional groups that is a part of the biopolymers contained in the shell of chestnut fruits. It was revealed that the acidity of the medium influences the degree of extraction of metal ions from aqueous solutions, while the temperature has little effect on the sorption capacity of the shell of chestnut fruits. It is shown that it is possible to increase the sorption characteristics of pollutants by chemical modification of the chestnut shells. It has been determined that sorption isotherms in most cases are more fully described by the Langmuir model, and the kinetics of the process in all cases obeys the pseudo-second order model.

1. Introduction

The aim of this work is to analyze and summarize the literature on the use of waste from the processing of buckwheat as sorption materials for the removal of pollutants from aqueous media. Nowadays, global scientific and industrial community intensively studies a new innovative direction in the field of environmental protection - the use of industrial waste [1, 2], lignocellulosic waste from agricultural production, as well as components and waste from the processing of wood coniferous and deciduous tree species as reagents to remove pollutants from aqueous media [3]. Ligno-, cellulose- and tannin-containing wastes from the processing of wood raw materials are of particular interest for the removal of pollutants of various origins from water bodies [4].
2. Materials and methods
Grinded bark, sawdust and leaves are effective adsorption materials (AM) for removing pollutants of various origins from natural and waste waters (WW). During the processing of tree fruits in industrial production, waste is also generated in the form of seeds, fruit peel and nutshells.

During the processing of chestnut fruits, waste forms in the shape of shells and internal partitions occur. Up to 20% of shell accounts for 100 kg of chestnuts, and from 2.5 to 4% of the inner shells adjacent to the fruit accounts for inner shells. Thus, from 100 kg of raw materials, the yield of peeled chestnuts is 75–78 kg.

Nutshells are studied as AM for the extraction of metal ions, dyes, oil products, and a number of other compounds from aqueous media.

This article summarizes information from literary sources on the use of chestnut shells as AM for the removal of pollutants of various natures from wastewater and natural waters.

In order to collect information, the authors used publications in leading Russian and foreign issues. During the research such methods as the methods of system analysis, comparative criteria for the effectiveness of scientific results were used.

The microstructure of the surface of the shell was studied using a high-resolution scanning electron microscope TESCAN MIRA 3 LMU.

3. Results and discussion

3.1. Basic information about chestnut
Chestnut (Lat. Castañea) is a small kind of trees of the Beech family (Fagaceae). Wood, bark and leaves contain from 6 to 14% of tannins. Chestnut has long been used as a fruit tree. Chestnut fruits are rich in carbohydrates and proteins, they are fried and baked, high-quality additives for flour. In addition confectionery products are obtained from them.

During the processing of chestnut fruits, waste forms in the shape of shells and internal partitions occur. The chestnut shell has no significant industrial and commercial use, which contributes to the emergence of a serious environmental problem.

3.2. Composition and possible uses of the chestnut shell
In the research work [5], it is indicated that during the processing of the chestnut a large amount of waste is generated, namely the inner shell, outer shell and leaves. The studies of chestnut by-products revealed a good profile of biologically active substances with antioxidant, anticarcinogenic, and cardioprotective properties. After valorization these agro-industrial wastes can be used by other industries, such as pharmaceutical, food or cosmetic.

The composition of the shells of chestnut fruits is determined in the research work [6], which is shown in Table 1.

| Ingredients               | Content, % |
|---------------------------|------------|
| Cellulose                 | 19.2       |
| Lignin                    | 29.15      |
| Soluble lignin            | **2.83**   |
| Total sugar content       | 33.82      |
| Of which monosaccharides  |            |
| - glucose                 | 19.23      |
| - galactose               | 2.98       |
| - xylose                  | 6.45       |
| - arabinose               | 2.52       |
| - mannose                 | 1.52       |
| Ash                       | 0.83       |
| Extracts:                 |            |
3.3. Sorption properties of chestnut nutshell components

One of the possible ways to use the chestnut nutshell is to use them as an AM for extracting pollutants from natural and waste waters. The analysis of literature shows that the largest number of publications is devoted to the adsorption of heavy metal ions (HMI). In this review, the information on the adsorption of HMI by the shell of the chestnut fruit is presented in the alphabetical order of metal ion.

A large number of publications are devoted to the study of adsorption of Cd\(^{2+}\) ions by the shell of chestnut fruits [7]. The experimental conditions and the obtained adsorption characteristics are shown in Table 2.

Table 2. Experimental conditions and adsorption characteristics for the adsorption of Cd (II) ions by the shells of chestnut fruits.

| Experimental conditions | Obtained adsorption characteristics | Note | Source |
|-------------------------|-------------------------------------|------|--------|
| $C_{\text{initial}}$ = 100–600 mg/dm\(^3\), pH = 1.0–7.0, sorbent dosage = 1–8 g/dm\(^3\), T = 30–70 °C, t = 0.5–7.0 h. | The maximum adsorption capacity is 51.28 mg/g. The maximum degree of removal of Cd ions is 98.9% | Discolored shell processed formaldehyde by cis-butenedioic acid, and irradiation. The adsorption isotherm is described by the Langmuir model ($R^2 > 0.9812$). Kinetics of pseudo-second order. | [7] |
| $C_{\text{initial}}$ = 20–100 mg/dm\(^3\), pH = 5.5–8.5, T = 10–40 °C, dosage = 10 g/dm\(^3\), t = 24 h. | The maximum sorption capacity is 4.75 mg/g. | Incomplete $3^3$ factorial level. The adsorption isotherm is described by the Freundlich model. | [8] |
| $C_{\text{initial}}$ = 250 mg/dm\(^3\), pH = 3–6, dosage = 10 g/dm\(^3\), t = 15 minutes | The maximum adsorption capacity is 14.7 mg/g. The degree of removal of Cd ions is 95%. | The adsorption isotherm is described by the Langmuir model ($R^2 > 0.998$). Adsorption kinetics corresponds to a pseudo-second order model. | [9] |

It is indicated that the adsorption of Cd\(^{2+}\) ions is carried out according to functional groups included in the biopolymers contained in the shells of chestnut fruits (-OH, C (O) OH, -NH and others).

3.3.1. Heavy metal ion adsorption

There are several publications on the adsorption of chromium ions by the fruit shells of chestnuts. In particular, the influence of the initial concentration of Cr (VI) ions – 100 or 150 mg/dm\(^3\), contact time – 2 and 6 hours and the pH of the medium – pH = 2 or 5 on the adsorption characteristics was studied. It was found that the best conditions for the adsorption of Cr (VI) ions were pH = 2 and a contact time of 6 h for both initial concentrations. It was established that adsorption isotherms are more accurately described by the Langmuir model [10].

Research work [11] also indicated that the removal efficiency of Cr\(^{6+}\) ions reaches 99.7% at pH = 2. It was also determined that the removal efficiency of Cr(VI) ions increased with increasing temperature in the range from 15 °C to 35 °C.

- cold water
- hot water
- 1 % solution of NaOH
- a mixture of ethanol, benzene and water

\[3.85 - 26.87 - 71.32 - 34.52\]
The model drains were cleaned of chromium ions using chrome chips impregnated with tannins from the chestnut shell as AMs. It was determined that the optimal pH of the medium for the efficient extraction of Cr (VI) ions was pH = 4, and the maximum adsorption capacity of sawdust was 42 mg/g. It was determined that the equilibrium conditions were reached after 2 hours of contact. It was revealed that the adsorption isotherm obeyed the Langmuir model, and the adsorption kinetics obeyed the pseudo-second order model [12].

There are several publications on the study of adsorption of Cu$^{2+}$ ions by the shells of chestnut fruits [13–15]. The experimental conditions and the obtained adsorption characteristics are shown in Table 3.

Table 3. Experimental conditions and adsorption characteristics for the adsorption of Cu(II) ions by the shells of chestnut fruits.

| Experimental conditions | Obtained adsorption characteristics | Note | Source |
|-------------------------|-------------------------------------|------|--------|
| Castaneamolissima, C$^{\text{initial}}$ 25–200 mg/dm$^3$, pH = 2–6, dosage 10 g/dm$^3$, T = 20–40 °C, t = 96 h, 120 rot/min | The adsorption capacity is 12.42 mg/g | Adsorption isotherm obeys Langmuir isotherm models. The kinetic data were found to obey the pseudo-second-order model. $\Delta G_0 = -5.875$, $-5.714$ and $5.442$ kJ/mol at T = 293, 303 and 313 K, $\Delta H_0 = -12.296$ kJ/mol, $\Delta S_0 = 21.534$ J/mol K. | [13] |
| Dynamic conditions. C$^{\text{initial}}$ = 20 mg/dm$^3$, pH = 6, particle size ~ 0.25 mm. | The maximum adsorption capacity is 10.94 mg/g | The adsorption process was endothermic. The process obeys the Thomas model. 96.1% was desorbed with 0.1 M HCl. | [14] |
| Insoluble pigment containing melanin from the chestnut shell. C$^{\text{initial}}$ = 10 mg/dm$^3$, t = 240 min | The maximum adsorption capacity is 33.2 mg/g | The kinetic adsorption behavior obeyed the pseudo-second-order rate law, and the equilibrium adsorption data were well described with both the Langmuir and the Freundlich isotherms | [15] |

In global literature there is little research works on the adsorption of Ni$^{2+}$ ions in the shells of chestnut fruits. The removal of Ni$^{2+}$ ions by some cheap AMs, including the shells of chestnut fruits, was studied [16]. The experiments were carried out at 25 °C with an initial concentration of a metal ion of 15 mg/dm$^3$ and the reaction time of 10–180 minutes at pH = 7 and a dosage of AM of 2–20 g/dm$^3$. It was found that the maximum adsorption capacity was 5.6 mg/g and the degree of extraction Ni (II) ions is 37.3%.

The adsorption of Pb(II) ions by Korean researchers was also studied [17]. The process was carried out at pH = 2–6 for 24 hours at a temperature of 20–40 °C under static conditions. The dosage of the shell was 5 g/dm$^3$. The constructed adsorption isotherms at temperatures of 20–40 °C were well described by the Langmuir model ($R^2 = 0.99$), and the kinetic dependences at the indicated temperatures at an initial concentration of Pb(II) ions from 10 to 40 mg/dm$^3$ obeyed a pseudo-second order model. It was determined that the maximum adsorption capacity of the chestnut shell was 31.3 mg/g.

There are practically no studies on the adsorption of Zn$^{2+}$ ions by the shells of chestnut fruits. Chinese authors conducted experiments on the use of chestnut shells as the AM of Zn (II) ions. The experimental results showed that when the concentration of the solution was 20 mg/dm$^3$, the pH was 6.03, the contact time was 120 minutes, and the dosage of the chestnut shell was 0.7 g, the degree of removal of zinc ions could reach 98.78% [18].

Several studies are carried out on the adsorption of the chestnut shell of several HMIs to compare adsorption activity. In particular, the influence of initial cation concentration, temperature and pH was investigated to optimize Pb$^{2+}$, Cu$^{2+}$ and Zn$^{2+}$ removal from aqueous solutions using acid formaldehyde.
pre-treated chestnut shell as adsorbent. The experiments were planned according to an incomplete 33 factorial experimental design. Under the optimal conditions selected, the metal ion adsorption equilibrium was satisfactorily described by the Langmuir isotherm model. The maximum pre-treated chestnut shell adsorption capacity was obtained for Pb$^{2+}$ ions 8.5 mg/g, for Cu$^{2+}$ ions 4.44 mg/g and for Zn$^{2+}$ ions 3.35 mg/g. A model that considered the effect of axial dispersion was successfully used to describe the fixed-bed adsorption behavior of Pb$^{2+}$, Cu$^{2+}$ and Zn$^{2+}$ ions at the flow rates essayed [19].

Korean authors studied the adsorption of Cd$^{2+}$, Cu$^{2+}$ and Pb$^{2+}$ ions with the skin of chestnut fruits. It was determined that the maximum adsorption capacity for these ions was in the following row: Pb$^{2+}$ (31.25 mg/g) > Cu$^{2+}$ (7.87 mg/g) > Cd$^{2+}$ (6.85 mg/g). It was shown that adsorption isotherms for the aforementioned ions of the chestnut shell were described by the Langmuir model ($R^2 = 0.99$) [20].

There are much more research works mainly published by Chinese researchers, in which the adsorption characteristics of the chestnut shell with respect to 4 metal ions are studied. In particular, the adsorption of Cd$^{2+}$, Cu$^{2+}$, Pb$^{2+}$ and Zn$^{2+}$ ions was studied. It was found that the maximum adsorption by ions for Cd$^{2+}$ ions occurred within 0.5 h, and for other ions after 1 h. It was determined that the temperature had little effect on the adsorption rate of Zn$^{2+}$ ions and increased with respect to Pb$^{2+}$ and Cd$^{2+}$ ions at 40 °C [31]. It was noted in the research work that the highest removal rate of Pb$^{2+}$, Cu$^{2+}$, and Zn$^{2+}$ ions was observed at pH = 5, and Cr$^{6+}$ ions at pH = 2 [21].

The adsorption of Cd$^{2+}$, Pb$^{2+}$, Cu$^{2+}$ and Zn$^{2+}$ ions from acidic solutions (pH = 2) by chestnut shells under static and dynamic conditions was studied. At an initial concentration of the mentioned HMIs of 30 mg/dm$^3$, the degree of their removal under static conditions after 1 h of contacting was 98.9% for Cd$^{2+}$ ions, 99.6% for Pb$^{2+}$ ions, 98.9% for Zn$^{2+}$ and 98.7 ions % – for Cu$^{2+}$ ions. The values of maximum adsorption capacity for these metal ions were: 3.2 mg/g for Cd$^{2+}$ ions, 90.8 mg/g for Pb$^{2+}$ ions, 27.3 mg/g for Zn$^{2+}$ ions and 52.4 mg/g by Cu$^{2+}$ ions.

Fourier transform infrared (FTIR) and X-ray photoelectron(XPS) spectroscopies showed that the functional groups involved in metal ions binding included carboxyl, hydroxyl, ether, alcoholic and amino groups.

Literary sources contain references to the work of Korean scientists on the adsorption of gold ions by the shells of chestnut fruits [22]. It is indicated that under the conditions of the experiment: pH = 2, temperature – 20 °C, when the initial concentration of Au (III) ions is 300 mg/dm$^3$, the maximum sorption capacity is 189 mg/g. It is shown that this value of the maximum capacity for gold ions is higher than that obtained by other researchers. The authors explain this fact by the presence of a large number of tannins in the composition of the chestnut fruit shell.

The adsorption of platinum ions by the chestnut shell was studied under the above mentioned conditions: pH = 2, temperature – 20 °C, stirrer speed – 120 rpm, dosage of sorption material – 2.5 g/dm$^3$. It was indicated that the maximum adsorption capacity, determined by the adsorption isotherm, was low and did not exceed 1.5 mg/g. The adsorption isotherm was adequately described by the Freindlich equation ($R^2 = 0.931$) [23].

### 3.3.2. Adsorption of organic compounds and dyes

Using the shell of chestnut fruits as an adsorption material to reduce the dyeability of solutions is not entirely advisable because the shell itself produces tannins and is used for dyeing, in particular, of fabrics and wool. Nevertheless, the possibility of extracting dyes of the Reactive Brilliant Red K-2G brand, Methylene blue [24] is indicated. It was determined that the maximum sorption capacity for the dye “Methylene blue” of the chestnut shell at 25 °C is 126.5 mg/g, and the adsorption isotherm was most fully described by the Langmuir model.

The adsorption of Auramine O, Basic Violet 14, Basic Blue 11, Rhodamine B, and Methylene Blue dyes with tannins isolated from the chestnut shell was studied. It was found that natural tannins were effective in removing dyes of the Basic Violet 14, Basic Blue 11, and Methylene Blue brands, and were ineffective in the other two dyes [25]. It was determined that the adsorption isotherms were well described by the Langmuir model.
The chestnut shell was also studied as an adsorption material for the removal of pesticides of the trade names Pirimicarb, Imidacloprid, Acetamiprid and Thiamethoxam under dynamic conditions\[26\]. It was determined that the adsorption capacity for these pesticides was: according to Thiamethoxam – 14.3 mg/g, Acetamiprid – 4.70 mg/g, Imidacloprid – 8.51 mg/g. It was found that treatment of the chestnut shell with citric acid increased the adsorption characteristics of these pesticides by 15%.

3.3.3. Horse-chestnut shell

In fact, besides species of the Chestnut kind, other plants from other families can be called “chestnuts”. For example, *Aesculus* - Horse Chestnut is a plant of the Soapberry family (*Sapindaceae*), common horse chestnut (*Aesculus hippocastanum* L.). Figure 1 shows a highly developed microrelief of the surface of the horse-chestnut shell.

![Figure 1](image1.png)

**Figure 1.** Microstructure: of chestnut skin surface (a, b); of the faults of peeled chestnut (c, d).

The shells of horse chestnut fruit were also investigated as AM HMIs. Thus, in particular, the adsorption of Cu$^{2+}$ ions by the green shell of an ordinary horse chestnut was studied. It was determined that the greatest extraction of copper ions at the initial concentration of the last 10–100 mg/dm$^3$ was observed at pH = 5, and the time to achieve adsorption equilibrium was 5 minutes. It was determined that the degree of extraction of Cu$^{2+}$ ions was 72.37–98.09%, depending on the process parameters. It was found that the adsorption isotherms were adequately described by the Freindlich model, and the kinetics of the process was described by a pseudo-second order model.

The adsorption of Cr (VI) ions with an initial concentration of 500–4000 mg/dm$^3$ was studied with varying process parameters of the shells of horse chestnut fruits. It was found that the highest sorption capacity (142.9 mg/g) was achieved at pH = 2 and contact time of 2 hours. It was found that the adsorption isotherm was most accurately described by the Langmuir model, and the kinetics of the process was described by a pseudo-second order model. The thermodynamic parameters of the process were determined, which indicate that the adsorption process was exothermic and was spontaneously.
Under dynamic conditions, the adsorption of Zn$^{2+}$ ions was studied using various AMs, including horse chestnut shells. It was found that the maximum adsorption capacity was 12.25 mg/g with an initial concentration of zinc ions of 97.5 mg/dm$^3$ at 25 °C.

4. Summary

Thus, the above mentioned materials summarize the literature on the use of grinded shells of chestnut fruits as an AM for the removal of metal ions, dyes and some organic compounds from aqueous media. It was determined that adsorption isotherms, in most cases, were more adequately described by the Langmuir model, and the kinetics of the process in all cases obeyed the pseudo-second order model. The values of adsorption indicators for the studied pollutants were given. It was shown that it was possible to increase the adsorption characteristics of the chestnut shell by chemical modification.

5. References

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