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Review of Pistachio (Pistacia) Shell Use to Remove Pollutants from Aqua Media

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Abstract: The paper summarizes literature data on using crushed pistachio shells (*Pistacia vera* L.) as sorption materials to remove ions of various metals, dyes, and antibiotics from aqueous media. It provides brief information on the amount of pistachio processing waste, its chemical composition, and its recycling methods. It gives the adsorption process parameters and the values of pistachio shell sorption parameters for the studied pollutants. It was shown that pistachio shells' sorption characteristics for various pollutants could be increased by chemical modification with various chemical reagents. The isotherms of pollutant adsorption with pistachio shells were found to be, in most cases, more accurately described by the Langmuir model, and the process kinetics to follow in most cases, the pseudo-second-order model. It is shown that the pistachio shell is a good precursor for activated carbons production, which can also be used for the adsorption of various pollutants from aqueous media.

Keywords: pistachio shells; metal ions; dyes; antibiotics; adsorption; modification; activated carbon.

1. Introduction

Currently, the world community is widely developing a new environmental protection area – the use of industrial and agricultural waste as reagents for removing pollutants from water environments [1-10]. Of particular interest are waste from processing tree biomass (sawdust, wood chips) and tree fruits (shells, pits, cones, etc.) [11-15].

It was shown that crushed bark [16], sawdust [17], and leaves [18] of trees are effective sorption materials to remove pollutants of various origins from natural and wastewater. In the industrial processing of tree fruits, waste is also generated in the form of seeds [19], fruit rind [20], and nut shells.

The latter has been studied as sorption materials to remove metal ions, dyes, petroleum products, and other compounds from aqueous media. However, the number of papers on using tree fruit processing waste is so large that many materials are considered in the corresponding reviews. In particular, we summarized the data using chestnut [21] and walnut [22] shells as sorption material to remove pollutants from aqueous media.

Pistachio is one of the trees whose nuts have been consumed by humans for over 2.5 thousand years. It is a small genus of evergreen or deciduous trees or shrubs in the

Anacardiaceae family, common in subtropical and partly in tropical areas. Pistachios are common in the Mediterranean, North-West Africa, Western, Central, and East Asia, on the Caspian and the Black Sea coasts of the Caucasus. Two species of pistachios are found in Central America, as well as in the south of Texas (USA).

The fruit of pistachios is a pseudomomeric drupe (pyrenarium), with a thin mesocarp and a hard stony endocarp. Pistachio is harvested in late July-early August. The world production of pistachio nuts in 2017 amounted to 1,115,066 tons. The major producers of pistachios (more than 100 thousand t/a) include Iran, Turkey, the United States, and China. Considering the fact that the ratio of the pistachio shell:nut is 0.45:1, the volume of formation of pistachio shells on a global scale is hundreds of thousands of tons per year.

The content of biogenic chemical elements in the pistachio shell was determined to be: carbon – 44.89%, hydrogen – 5.66%, nitrogen – 0.53%, sulfur – 0.97%, oxygen – 47.95% [23]. According to various sources, cellulose content is 38.1-54.0%, hemicellulose – 15.2-31.4%, lignin – 25.2-29.4% [24-26]. According to research, the main component of pistachio (Pistacia vera L.) by-product with the highest concentration of bioactive substances (such as polyphenols, tocopherols, dietary fibers, essential oils, and unsaturated fatty acids) with antioxidant properties and health-promoting effects is pistachio hull. [27]. Besides, certain metals and metalloids were identified in the pistachio shells, such as Sn, Ca, K, Na, Mg, Fe, Ni, Mn, Cu, Zn, and Si. Their average content (mg/kg) was determined as follows: 1.63, 424.90, 1646, 155.60, 296.2, 420, 31.52, 6.27, 5.26, 1.45 and 153.46, respectively [28].

However, pistachio shells recycling is a certain challenge. There is information in the world literature about using pistachio shells for energy production by burning [23] or pyrolysis [29,30]. The possibility of using pistachio shells to produce nanocellulose is shown [31,32]. The extract from the pistachio shell can be used as a coloring agent [33]. The use of small-sized particles of pistachio fruits as a filler for elastomeric compositions [34] and plastics [35] has been proposed.

One of the ways to use pistachio shells is as sorption materials to remove various pollutants from aqueous media.

2. Heavy Metal Ions

The presence in the pistachio shell of many biogenic compounds with various functional groupings is supposed to contribute to the high adsorption properties of the latter in respect of ions of various metals and metalloids. The data on the use of pistachio shells as a sorption material to remove metal ions is given in Table 1.

Table 1. Experiment conditions and metal ions adsorption characteristics of native and modified pistachio shells.

Metal ion	Experiment conditions	Adsorption characteristics	Note	Source
Cr(VI)	pH = 2–8, DS = 0.5–8 g/dm^3, C_0 = 50–200 mg/dm^3, T = 5–50 °C t = 1–60 min.	A_{Max} = 116.3 mg/g. Over 99% of chromium from solutions containing 50–200 mg/dm^3 of Cr(VI) ions at a pH = 2 and an adsorbent concentration of 5 g/dm^3 after 60 min.	Kinetic and isotherm modeling studies demonstrated that the experimental data best fit a pseudo-second-order and Langmuir model, respectively	[36]
Cu^{2+}	Dynamic conditions. 100 sm^3 of 25 mg/dm^3 Cu^{2+} solution was poured in every 250 dm^3 stoppered conical flask and adjusted to initial pH in the range from 3 to < 6.	A_{Max} = 33.25mg/g.	Thomas's model performed well for the selected operating range (R^2 = 0.9431)	[37]

Metal ion	Experiment conditions	Adsorption characteristics	Note	Source
Hg^{2+}	pH = 3–9, DS = 0.1–1g/dm^3, C_0 = 50–200mg/dm^3, T = 30 °C t = 10–90min.	A_{Max} = 48.78mg/g	Pseudo-second-order model was the best model representing the adsorption of Hg^{2+} ions. Activation energy 10.26 kJ/ mol.	[38]
Ni^{2+}	pH = 2–10, DS = 5–30 g/dm^3, C_0 = 20–600 mg/dm^3, T = 25 °C t = 5–180 min., 250 rpm	A_{Max} = 14,1 mg/g	The Freundlich isotherm best fitted the experimental data (R^2 = 0.9654)	[39]
Pb^{2+}	pH = 2–11, DS = 0.5-4.0 g/dm^3, C_0 = 10–100 mg/dm^3, T = 20-60 °C, t = 20–60 min., 250 rpm.	When the initial Pb^{2+} ions concentration was increased from 5 mg/dm^3 to 100 mg/dm^3, the loading capacity increased from 1 mg/g to 19 mg/g of the dried adsorbent mass.	The adsorption process obeys Freundlich isotherm	[40] [41] [42]
Pb^{2+}	pH = 3–9, DS = 0.5-2.0 g/dm^3, C_0 = 10–150 mg/dm^3, T = 20-60 °C, t = 5-30 min., 140 rpm.	reduction of 80.71 % Pb^{2+} ions removal	Better linearity was exhibited by Freundlich isotherm	[43]
Pb^{2+}	Pistachio-hard shell-graft-thiosemicarbazone acetophenone composite matrix. pH = 2–10, DS = 50-200 mg/dm^3, C_0 = 25–150 mg/dm^3, T = 298-313 K, t = 2-10 hrs., 180 rpm.	The optimum dosage of the modified adsorbent was 0.165 g for the adsorption process at pH = 5. The removal percentage of Pb^{2+} increased from (62.54 to 92.55 %), (42.71 to 76.0 %) and (32.49 to 60.0 %) at 25, 40 and 100 mg/dm^3 initial lead concentration, respectively	Freundlich isotherm model. Kinetic data were best described by the pseudo-second-order model	[44]
U(VI)	From seawater. pH = 8, DS = 60 g/dm^3, C_0 = 15 and 100 ppm, T = 22 °C, t = 1-270 min., 300 rpm.	A_{Max} = 185.45 μg/g of the shell at 15 ppm, and A_{Max} = 370.37 μg/g of the shell at 100 ppm.	The mechanism followed pseudo-second-order and intraparticle kinetics models, and the shell was demonstrated to be a Freundlich isotherm	[45]
Zn^{2+}	pH = 2–8, DS = 2-10 g/dm^3, C_0 = 367.25 mg/dm^3, T = 25 °C, t = 5-120 min., 200 rpm.	The uptake of Zn^{2+} ions increases from 97.9 % at 8 g/dm^3 to 98.1 % at 10 g/dm^3.	The adsorption process follows the pseudo-second-order kinetic model. ΔG^0 = -45.998 – -48.380 KJ/mol (293-323 K), ΔH^0 = -22.734 KJ/mol, ΔS^0 = 79.398 J/mol·K	[46]

C0 is the initial concentration of pollutants in solutions, DS is the dosage of the sorption material, T is the experiment temperature, t is the adsorption time, and Amax is the maximum adsorption capacity.

Some publications are devoted to studying the extraction of several metal ions from aqueous media by pistachio shells. Thus, the powder of native pistachio shells was used to remove Cd^{2+} and Pb^{2+} ions from simulated solutions. The values of the maximum sorption capacity calculated according to the Langmuir equation were 14.9 and 142mg/g, respectively, for Cd^{2+} and Pb^{2+} ions. The kinetic experiments showed that Cd(II) and Pb(II) biosorption on the adsorbent is rapid, and maximum biosorption capacities were achieved in 2 h. The adsorption isotherms showed that Cd(II) and Pb(II) affinity to the adsorbent increased with pH. The equilibrium adsorption of Cd(II) and Pb(II) was satisfactorily described by the Sips isotherm. The time-dependent biosorption of Cd(II) and Pb(II) onto the adsorbent was well described by both the pseudo-first-order and the pseudo-second-order models [47].

Using a pistachio shell as a biosorbent, the biosorption of Co2+ and Pb2+ ions from an aqueous solution is investigated. The aqueous solution's pH, the metal's initial concentration (C0), the biosorbent dosage (DB), and the temperature all affect how much metal is removed

(T).It was discovered that Pb2+ ion adsorption is what produces the actual maximum results: when pH is 5, C0 is 11.75 mg/dm3, DB is 0.8 g/ dm^3, and the temperature is 25 °C, the removal efficiency is 85.3%. When pH is 5, C0 is 5 mg/ dm^3, DB is 0.76 g/ dm^3, and T is 25 °C, removal efficiency is 74.4%. [48].

Investigate the possibility of using a soft pistachio shell to eliminate lead (II) and mercury (II) ions from aqueous solutions as a low-cost, efficient substitute adsorbent. The initial concentration of these ions was 10mg/dm^3. Obtained results gave a removal of 90.9 % for Pb^{2+} ions in the pH = 6, exposure time of 80 min at 25°C. Maximum removal of Hg^{2+} ions of 91.5 % was obtained in pH = 6 at 25 °C after an exposure time of 100 min. The Langmuir and Freundlich equations for describing sorption equilibrium were applied, and the results indicate that Freundlich well described the process. Also, adsorption kinetics data were modeled using the pseudo-first and pseudo-second order; it was found that the second-order model describes both ions' uptake better [49].

The potential of powdered pistachio hull (PHP) for the co-adsorption of Cr(VI) and cyanide ions from electroplating wastewater was compared. The results of dynamic adsorption experiments indicated that the complete and simultaneous removal of Cr(VI) and cyanide from wastewater were achieved with 2 g/dm^3 of PHP after 60 min of contact. The maximum capacity of PHP for the co-adsorption of Cr(VI) and cyanide-ions were 117.6 and 151.5 mg/g, respectively. PHP's adsorption of Cr(VI) and cyanide-ions followed pseudo-second-order kinetics, and the equilibrium adsorption data best fit the Langmuir isotherm [50].

Pistachio shells have also been studied for their ability to soften water and remove Ca^{2+} and Mg^{2+} ions. The optimum adsorption conditions are at pH = 8, and the weight of the pistachio shell is 1.5 g/dm^3. The adsorption capacity of Ca^{2+} and Mg^{2+} ions is 2.41 and 2.19 mg/g, respectively [51].

Peanut, hazelnut, pistachio, walnut, and almond shells were studied for the ability to remove Cd^{2+}, Pb^{2+}, and Hg^{2+} ions from contaminated water. The sorbents dosage was 0.5g/dm^3. Ultrapure water mono-element spiked with 50 µg/dm^3 of Hg^{2+} ions, 200 µg/ dm^3 of Cd^{2+} ions, and 1000 µg/dm^3 of Pb^{2+} ions. The degree of metal ions removal by pistachio shells was: for Cd^{2+} ions-94%, for Pb^{2+} ions – 96%, and for Hg^{2+} ions – 72%. The most effective sorbent, hazelnut shells, were determined and tested in mono- and multi-contaminated mineral water. [52].

3. Dyes and other organic compounds

Pistachio shells were also studied as a sorption material to extract various dyes from aqueous media (Table 2).

Table 2. Experiment conditions and dye adsorption characteristics of native and modified pistachio shells.

Dye	Adsorption characteristics	Note	Source
Reactive red 120	A_{Max} = 324.9mg/g	Langmuir model. The logistic model was well correlated with the initial dye concentration.	[53]
Reactive Red 45	A_{Max} = 35.336mg/g.	Freundlich isotherm is the best fitted for adsorption (R^2=0.9969)	[54]
Remazol Red	A_{Max} = 108mg/g at 20°C	Freundlich isotherm is the best fitted for adsorption (R^2>0.99)	[55]
Reactive Red 238	A_{Max} = 109.535mg/g.	The results revealed that the biosorption data were best fitted by the pseudo-second-order and Sips models. E = 0.164 kJ/mol, $\Delta G^0 = -5.184$ kJ/mol for 30°C	[56]
Methylene blue	A_{max} = 389 and 602 mg/g for temperatures between 20 and 50 ◦C, respectively	The kinetic analysis showed the pseudo-second-order model. Langmuir model.	[57]

Dye	Adsorption characteristics	Note	Source
Methylene blue	A_{max} = 16.74 mg/g at 308 K, 120 rpm, 24 h.	Isotherm studies revealed the applicability of the Sips model. Pseudo second order kinetics model.	[58]
Basic blue 41	A_{max} = 41.77mg/g.	The results of modeling studies proved that Langmuir isotherms and pseudo-second-order kinetics were the best-represented adsorption kinetics and isotherm data.	[59]
Direct blue 71	A_{max} = 90.48mg/g.	The Freundlich isotherm (R^2 = 0.9912) model. The pseudo-second-order model accurately captured the adsorption kinetics. The adsorption process is a spontaneous and endothermic process.	[60]
Cibacron blue	A_{max} = 1.63 mg/g (pistachio shell), A_{max} = 4.53 mg/g (treated pistachio shell).	Equilibrium adsorption isotherm and kinetic studies showed that The pseudo-second-order kinetic model and the Langmuir isotherm provided good fits to the experimental data.	[61]
Basic violet 14 (Fuchsin)	Removal of 99.71% of BV14 with an adsorption capacity of 118.2 mg/g (pH=12, DS=100 mg/50dm^3, C_0=250 ppm, 20min	The kinetic analysis showed the pseudo-second-order model. Langmuir model.	[62]
Crystal violet	Removal of 93.6 % at pH = 6, initial dye concentration 10 mg/dm^3, contact time 360 min and adsorbent dose 4 g/dm^3	Optimization of the process by the Taguchi method.	[63]
Acid violet 17	Dynamic conditions. A_{max} = 13.66 mg/g at 2 cm bed height, 6 sm^3/min flow rate, and 40 mg/dm^3 initial concentration	Modification with H_2SO_4. The Thomas model is more suitable for the description of breakthrough curves.	[64]
Acid Violet 17	A_{max} = 26.46 mg/g at the initial dye concentration of 160 mg/dm^3	Modification with H_2SO_4. The adsorption follows both the Temkin and the Langmuir isotherms. An estimated pseudo-second-order kinetic was found.	[65]
Chrysoidine Y	A_{max} = 79.37 mg/g at pH = 5, at 2.5 hours contact time, at 1.0-1.25 g/100 dm^3 adsorbent dosage	Pseudo-second-order model provided the best correlation, and the rate-limiting step might be chemical sorption. The Freundlich isotherm was feasible to identify the adsorption process	[66]
Remazol Brillant Blue R	A_{max} = 16.739mg/g.	The data fit the Langmuir Isotherm (R^2=0.9989). ΔG = 2.696 kJ/mol.	[67]

And $_{max}$ is the maximum adsorption capacity

The data on the maximum sorption capacity for dyes given in Table 1 are not comparable since the experiments were carried out under different conditions. The sorption characteristics can only be compared if the experiments were carried out under equal conditions.

In particular, the removal of Acid Red 42 (AR42) as an anionic dye and Basic Red 18 (BR18) as an acid dye from an aqueous solution by adsorption is investigated with pistachio hull as industrial waste. Both isotherm and kinetic models were applied to remove dyes in a batch system. The batch adsorption data were suitable for the Freundlich isotherm model (R2=0.9954) for BR18 and Langmuir isotherm model (R2=0.9927) for AR42. Data fitted a pseudo-second-order kinetic model for this study. It was found that the maximum dye sorption capacity determined from the Langmuir model equation for AR42 was 85.6mg/g, for BR18 – 127.8mg/g [68].

Pist pistachio shells as an adsorbent for the removal of Brillant Blue and K-RED 198, Methylene Orange, and Methylene Blue dyes were investigated. The commonly used isotherm models were applied for data obtained from further batch studies. Dye removal capacity is 65% for Brillant Blue and 73 % for KRED 198. Methylene Orange and Methylene Blue were removed poorly for the pistachio shell. Freundlich isotherm model was found to be the best-fitted one, and based on the Freundlich isotherm model, adsorption capacities were 4.04 mg/g for Brillant Blue and 4.64 mg/g for K-RED 198 at pistachio shells. Kinetic examinations were also carried out for two dyes tested, and it was found that adsorption kinetic was best described by a pseudo-first-order kinetic model [69].

In addition to dyes, pistachio shells have been studied as sorption materials to remove certain antibiotics from aqueous media. In particular, It has been investigated how to remove the veterinary fluoroquinolone antibiotic Sarafloxacin (SARA) from wastewater with pistachio hull. It was found that the maximum sorption capacity for the above antibiotic was 7.61mg/g [78].

They investigated the efficiency of the pistachio shell coated with ZnO nanoparticles (CPS) for tetracycline sequestration from wastewater. An isotherm model reading revealed that the results of the experimental work were a good fit with the Freundlich model. A maximum adsorption capacity (95.06 mg/g) was attained at 70 ppm initial concentration under the ideal conditions (pH = 4, dosage = 0.08 g/mL, particle size = 87 mm, shaking speed = 150 rpm, 25° C). The pseudo-second with a high coefficient of determination and a closer match to the experimental uptake, the pseudo-second-order model best describes the kinetics of tetracycline adsorption onto CPS. Intraparticle diffusion is not the primary mechanism, however. Tetracycline adsorption onto CPS was exothermic and spontaneous, according to thermodynamic characteristics. [71].

Following on from the previous section, the simultaneous adsorption of tetracycline (TEC), amoxicillin (AMO), and ciprofloxacin (CIP) from an aqueous solution were investigated using pistachio shell powder that had been coated with ZnO nanoparticles (CPS).The adsorption capacity of the pistachio shell was dramatically enhanced after post-coating with ZnO nanoparticles on various surfaces and structural elements. The correlation of the kinetic data by a pseudo-second-order model was successful for three antibiotics. The AMO isotherm curves had a better fit with the Langmuir model. The TEC and CIP isotherm measurements and the Freundlich model showed excellent agreement. Additionally, its spontaneous and exothermic nature characterized the three antibiotics' adsorption process onto CPS. According to the findings, the AMO, CIP, and TEC reactions on the homogeneous and heterogeneous sites of CPS surfaces have controlled chemical adsorption. The maximum adsorption capacities for TEC (98.717 mg/g) and CIP (92.450 mg/g) were shown by the CPS, followed by AMO (132.240 mg/g). [72].

4. Activated Charcoal from Pistachio Shells

One of pistachio shells applications is the production of activated carbons and carbonizates from them [73-75] and using the latter in various productions, including removing various pollutants from aqueous media.

In the world literature, activated carbons produced from pistachio shells were studied to remove Pb 2+ ions from water [76-80]. It was found that the maximum sorption capacity for these ions was from 0.17 to 24mg/g. In addition, activated charcoal made from pistachio shells was used for the adsorption of ions of As(III) and As(V) [77], Cd^{2+} [79], Cu^{2+} [79-81], Zn^{2+} [79], Pt^{4+} [82], U(VI) [83, 84]. It was determined that the sorption capacity for these ions did not exceed 50mg/g. The use of activated carbons from pistachio shells to extract U(VI) ions is much more effective, according to which the maximum sorption capacity reaches 335 mg/g [84].

Much more effective is the use of activated carbons produced by the carbonization of pistachio shells to remove dyes from aqueous media. In particular, there was a study of the adsorption of Methylene blue (Amax = 155.6 mg/g) [85], Aid Violet 17 (125 mg/g) [86], Crystal Violet (196.08 mg/g) [87], Methyl red" (144-264 mg/g) [88] dyes. It is obvious that the maximum sorption capacity of the activated carbons for dyes exceeds 100mg/g.

5. Conclusions

This review summarizes literature data on the use of pistachio (Pistacia vera L.) shells as a sorption material to remove ions of various metals (Ca^{2+}, Cd^{2+}, Co^{2+}, Cr(VI), Cu^{2+}, Hg^{2+}, Ni^{2+}, Pb^{2+}, U(VI), Zn^{2+}), dyes, and some antibiotics from aqueous environments. The paper provides brief information on the amount of pistachio processing waste, its chemical composition, and its recycling methods. It gives the adsorption process parameters and the values of sorption parameters for the studied pollutants. It was shown that pistachio shells' sorption characteristics for various pollutants could be increased by chemical modification with various chemicals. It was determined that the Langmuir model more accurately describes the pollutants' adsorption isotherms in most cases. The pseudo-second-order model is typically used to describe the process's dynamics. It is shown that pistachio shells are good precursors for activated carbons production, which are also promising sorbents for the extraction of metal ions and dyes from simulated solutions and wastewater.

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Conflicts of Interest

The authors declare no conflict of interest.

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