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Waste to Wealth: Sustainable Valorization of Walnut Shells into High-Value Bio-Based Materials

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Abstract:
Sustainable management of agricultural wastes has gained more and more attention in the world particularly in the production of value added products that are renewable and rich in carbon. Moreover there is the necessity to offer low cost and lower energy alternatives to active carbon and removal of contaminants in water. The walnut shell is an organic abrasive of hard nature and is environmentally and cost effective. Ash made of walnut shells, which is rich in alkali and alkaline earth elements has potential as a catalyst in transesterification reaction. Besides walnut shells can be converted to biochar through pyrolysis and this product is a low cost adsorbent that is useful in the removal of arsenic. Iron loaded biochar (ILB) which is a product of waste walnut shells has a significant arsenic adsorption capacity. Being a renewable resource, the walnut shell biomass has a broad spectrum of applications in bio based products such as biofuels bioenergy biochemicals and biomaterials. A review will be done on the different thermochemical conversion processes with emphasis on liquefaction of wood and how biomass composition affects the liquefaction process and chemical properties of the weakening biomass liquefied product. Walnut shells (WS) are a good example of hazardous material removal bio sorbents owing to their extraordinary absorbency and bio sorbent properties. WS powder has also antioxidant compounds as well as useful substances such as charcoal and pyroligneous acids. In addition to bio sorbent walnut shell is also handy in reinforcement filler to thermoplastic composites especially in outdoor applications where the component has to be very dimensional stable. Their versatility and potential are evidenced by its use in various industries, which show the flexibility and innovativeness of the walnut shell.

Key words: Walnut Shell, Waste Valorization, Biochar, Biomass, Biosorbent, Pyrolysis, Arsenic Removal, Biodiesel Catalyst, Activated Carbon, Sustainable Bio Based Materials

Introduction

Walnut belongs to the famous family of Juglandaceae. Fruit is made of a thin layer known as skin or seed coat, and an outer layer known as husk made of green shell which is organic and

biodegradable (Jahanban-Esfahlan et al., 2019). walnut shell is an organic, biodegradable, raw material (Shalpykov K et al., 2017).
J.regia has long been introduced to a far greater region, because of its special nutritional value, and its great commercial value. Commercial growing of walnuts was done on 647.497 hectares of land in 48 countries in 2016. Following 3,763.725 metric tons (in shell) of walnuts in the world in 2016, it is estimated that the total value of the production worldwide is more than USD 14 billion. China and the United States produced 47 and 31 percent of all the produced walnuts across the globe correspondingly (Shigavea and Darr., 2020).
The ash that is generated after the shell of walnut is burned can be used as a catalyst, adsorbent or catalyst when preparing biodiesel (Alba-Rubio et al., 2012). Microwave-pyrolyzed walnut shells through iron loading have also shown the potential of the material to be used as a low-cost adsorbent to remove arsenic (Miladinovic et al., 2020). One of the most frequent thermochemical conversion processes is liquefaction of wood in the presence of organic reagents, which are currently being studied to understand the influence of the biomass content on the liquefaction characteristics and properties (Antonovic et al., 2019). Activated carbon and carbon nanotubes are made of the walnut shells because of their huge surface area and porosity, and this could be used to clean water (Ghasemi et al., 2015).
Lignin was also exploited in lignin-based glues, activated carbon produced out of walnut shells (Wang et al., 2022). Fire ceramic bricks were made by burning clay using a waste walnut shell. Walnuts may be utilized as a replacement of less conservative materials or synthetic fillers in the reinforcement of different types of polymer composites (Shejkar et al., 2020) Walnut shell is often used to cleanse the soil of pesticide residues. (Liu et al., 2022) Starting with the use of walnut shells to create a series of pharmaceuticals with diverse therapeutic and preventive properties by releasing desirable residual biologically active chemicals (Harini et al., 2018).
Because hygroscopic components are lower in the quantities of a wood, and the quantities of hydrophobic components are larger in the walnut shells than in the wood, composite materials that are made of polymer that uses filler of the walnut shells have great business prospects in outdoor products that need a high level of resistance against the environment such as flooring or fencing (Shejkar et al., 2020).

Chemical Constituents of Walnut Shell

Ash	3.4%	Abdulwahid et al., 2023
Lignin	29.9%	Queirós et al., 2020
cellulose	30.4%	Domingos et al., 2022
Hemicellulose	24.9%	Domingos et al., 2022
Protein	2.4 to 2.5 %	Kizatova et al., 2022
Fat	0.7 to 0.9 %	Kizatova et a., 2022
minerals	1.5-2%	Queirós et al., 2020

Bioactive Potential of Walnut Shell

Lignin cellulose oleic, and palmitic acids are some of the important bioactive compounds in WS (Fordos et al., 2023). walnut shell is an organic and biodegradable raw material. Firstly processed shells are unique raw materials of cosmetic, pharmaceutical, food, fuel industries. Walnut shell is perfectly safe both to the surroundings and the human health. Ground walnut shell is a homogeneous natural powder which does not harbor any allergies does not retain water hence does not support the development of mold and microbes. It is an excellent raw material when it comes to the manufacturing of body care products the manufacture of washing pastes scrubs of all calibrations (Shalpykov K et al., 2017).

The application of walnut shells as raw materials to acquire drugs of different therapeutic and preventive properties depending on the release of residual valuable biologically active substances and the investigation of possibilities of developing a model of the waste free technology of the processing of walnut and the study of groups of biologically active substances of the shell and the viability of the economic feasibility of such researches is topical and promising branch of scientific research. (Sultanova et al., 2022)

Walnut Shell Valorization:

Catalyst for Biodiesel

In addition to energy (heat/electricity) the ash is a solid waste product resulting on burning walnut shells that may have a limited use as other ashes, e.g. as a catalyst a supporting material to other catalytical species or as an adsorbent. Activated carbon is prepared using the shell of the walnut, and was evaluated as an adsorbent of volatile organic compounds and supporting La and Ca used to prepare biodiesel. Yet the catalytic activity of walnut shell ash has not been experimented so far in transesterification reactions even though their chemical composition (alkali and alkali earth predominant elements) indicates that it may be used as a catalyst. Moreover the ash of walnut shell has not been fully described as a solid catalyst. (Alba-Rubio et al., 2012)

Walnut Shell Biochar for Sustainable Arsenic Removal

One of the typical ways of utilizing this waste rich in carbon is through combustion which has adverse negative effects on the environment. Prior to this study the use of walnut shells to generate a microwave pyrolyzed iron-loaded biochar and assess its applicability as a low cost adsorbent to remove arsenic was already done (Miladinovic et al., 2020). Carbo Culture that is located in the Central Valley of California has provided the walnut shell biochar. The pyrolysis oven was heated to 10 bar of the shells (Ndoun et al., 2023).

Microwave-aided pyrolysis for arsenic removal

Pyrolysis is a thermochemical process which does not require oxygen and transforms biomass into charcoal. Typically it involves long traditional heating which increases the price of the produced biochar. Microwave aided pyrolysis is increasingly becoming the method of manufacturing biochar giving it higher surface area than the traditional heating method as well as being an energy efficient and clean method of heating. The removal of Arsenic (V) has occurred when iron oxide coating on a range of porous precursors as alumina activated carbon cloth fibers aerogels, fly ash, and membranes is used (Mohamed et al., 2016).

Walnut shell was used to produce iron loaded biochar (ILB) by means of microwave pyrolysis. It was microwave assisted pyrolysis of $FeCl_3$ treated walnut shells that was conducted using 20 minutes of input power of 800 W to produce the iron loaded biochar. The arsenic adsorption capacity of the resultant product at 25 degC is 1.91 mg/g (Duan et al., 2017).

Walnut shell Liquefaction

The new technologies in the efficient use of biomass will make the creation of the environmentally friendly bio based goods such as biofuels bioenergy biochemicals and biomaterials possible (Misra et al., 2014).

The most significant interest was taken in the thermochemical conversion methods of wood liquefaction using some organic reagents. Research has been done on how the chemical structure of biomass influences the behavior of liquefaction and the chemical properties of liquefied biomass. The most interesting methods that were used in the previous study and reported literature refer to the liquid reaction of phenols and polyhydric alcohols (Antonovic et al., 2019).

Pyro ligneous acid production from walnut shell

The slow pyrolysis of plant biomass converts the biomass into pyroligneous acid. It is acidic, dark brown or yellowish brown in color (Mathew and Zakaria, Z. A. 2015). Pyrolysis vapors condense to produce this highly oxygenated aqueous liquid fraction, which is a complex (Ma and C et al., 2013).

Antioxidant activity of pyro ligneous acid from walnut shell

The pyro ligneous acid was assessed by a standard test as follows to 0.5 ml of the pyro ligneous acid 0.1 ml of 0.5 M NaOH was added and the mixture was swirled thoroughly to form a bicarbonate. 0.1 ml of a 1.2 M NaHCl solution was added to the bicarbonate and the mixture was swirled thoroughly to form an acid solution.

Walnut shells pyroligneous acids were also produced at different temperatures and their antioxidant ability was explored that can be used as antioxidants. The antioxidant activity of pyroligneous acids extracted by using shells of the walnut was studied by the following metrics which comprised such as DPPH (2,2-Diphenyl-1-picrylhydrazyl) free radical scavenging capacity hydroxyl free radical scavenging capacity superoxide anion radical scavenging capacity reducing power and anti-lipid peroxidation ability (Wei et al., 2010).

Carbon nanotubes from waste walnut shells

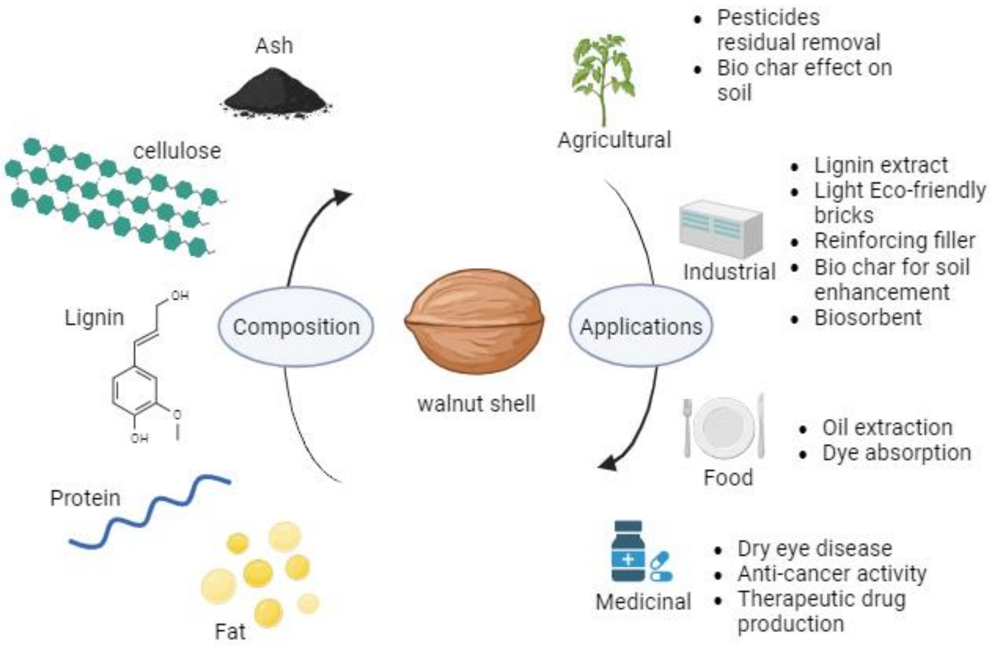
The Carbon nanotubes have been made extensively using walnut as well as other shells that are of a plant. Activated carbon has been made using a chemical activation process using walnut shells. The artificial activation of carbon was huge in its surface area and extremely porous. The activated carbon produced using walnut shells was found to have good absorption of iron and chromium in aqueous solution, which implies that the sample could be applicable to the purification of contaminated waters (Ghasemi et al., 2015).

The activated carbon of walnut shell is applied in treatment of industrial wastewater. A chemical activation process was done to produce activated carbon. Zinc chloride treatment was used to get activated carbon out of the shell and the shell was washed and left to dry. The activated carbon was a good heavy metal adsorbent depending on the pH Jiang and the duration of time the sample was to be kept in contact with the activated carbon. (Jiang and Y et al., 2017)

The Walnut shells composite prepared and used as a CNT absorber have achieved success in adsorption of heavy metals on aqueous solutions. Walnut shell and CNT composite had the highest lead adsorption (Saadat et al., 2014).

Applications Across Various Fields

The crushed walnut shell is a universal environmentally safe raw material with distinctive physical properties and chemical properties which has numerous applications in different spheres of industries (Ospanov et al., 2019).



Usage of walnut shells in different areas is demonstrated in figure.1.

Industrial applications

Lignin extract

The hulls and walnut shells are highly value added products that find application in food industry and other industries. As an example Santogo et al. 2000 used two extraction sequences to extract lignin of a Walnut shell. Auto hydrolysis delignification solid cleaning and precipitation are the processes that promote the production of lignin. Activated carbon made out of walnut shells and lignin made adhesives were highly used with lignin (Wang et al., 2022). The processing industry of walnuts produces around 400g of shells in the form of by products per kilogram of processed walnuts; these by products can be disposed of or burnt to generate fuel (Jahanban- Esfahlan et al., 2020). Nevertheless due to their large mass concentration of lignin (approximately half of the total), such wastes can be turned into lignin by means of a circular economy approach (Jahanban-Esfahlan et al., 2020) Precipitation delignification and autohydrolysis and solid cleaning are some of the processes involved in the production of lignin (Santigo et al., 2022).

Light Eco-friendly bricks

The process of doping agricultural waste with burned bricks is important in the production of light colored environmentally friendly bricks with a better thermal shielding. Burned clay wastage walnut shell was added to make the fire ceramic bricks. The findings indicated that there was an enhancement of specific heat capacity and absorption of water. The higher the WS content the higher the porosity and water absorbing properties were associated with it. Thermal conductivity and linear shrinkage decreased with the increase in the content of walnuts at higher temperatures 1100 950 C at C respectively. (Barnabas et al. 2023)
walnut shell in mortar instead of part of the sand, and the results showed that the material has good strength and thermal insulation properties as well as produces a more eco friendly mortar (Abdulwahid and Abdullah., 2020) Rigid polyurethane foams reinforced with walnut shell and its derivatives were also synthesized (Czlonka et al. 2020a, 2020b). However no use of this

substance as pore-forming agent of burnt clay bricks has been done. Besides identifying the optimal amount of walnut shell that would be allowed to form the development of masonry bricks at a fire temperature of 950 degC and 1100 degC, the current study also examined the usage of the walnut shell as pore forming in the production of the sustainable and energy efficient bricks to be used in construction. (Barnabas et al., 2023)

Walnut shell reinforcing filler

Thermoplastic composites are able to achieve a competitive advantage when walnut shell is used as a reinforcing filler in outdoor applications of such deck flooring or siding where dimensional stability is paramount to the success of the construction. These advantages are an increase in concentrations of hydrophobic components lignin and extractives and reduced concentrations of hygroscopic elements (cellulose and hemicellulose) in the cell walls of walnut shells compared to wood. (Ashori et al. 2010)

The polymeric structure of the composites was interwoven with the mechanical and thermo-mechanical properties to be altered by the inclusion of a ground walnut shell into the composite. Composites based on natural fillers have a lower tensile and impact strength and a significantly higher stiffness and hardness. (Salasinska et al., 2022

The powdered walnut shell improves different properties of the polymeric composites. Walnut is applicable in synthetic fillers substitutes or more conservative elements when reinforcing any type of polymer composite. (Shejkar et al., 2020)

Walnut shell biochar for Soil enhancement and Renewable energy generation

The application of biochar as a black carbon product made by thermal or hydrothermal treatment of biomass would be strongly recommended as a soil supplement because it can reduce greenhouse gas emissions and improve the soil fertility through retention of more nutrients and moisture (Tang et al., 2013).Walnut shells are considered as biomass residues during the production of biochar. Walnut shell biochar has a lot of opportunities to be used as an efficient fuel in homes and commercial furnaces to generate energy (Alfattani et al., 2021).

Use of walnut shell as a bio-sorbent for the removal of hazardous material

The walnut shell powder is highly absorbent and reinforcing (Ding et al., 2013). WS has also been used in addition to production of ACs and removal of HM ions and oils due to special mechanical properties, low ash level, and bulk density (Sarsari et al., 2016).

Bio absorption of Cr (VI) Ions using walnut shell

The bio absorption of Cr (VI) ions in aqueous solutions and the electroplating of a wastewater with walnut shell powder in a batch bio absorption procedure. The wastewater sample electroplating was effectively treated by the bio absorption procedure in which the concentration of chromium (VI) ions, organic compounds, and COD were reduced effectively. Walnut shell powder has also been believed to be an effective, cheap and environmentally acceptable bio absorbent on the basis of its sorption capacity to remove Cr (VI) ions in aqueous solution and the electroplating effluent (Farhan et al., 2012).

Table 3. Maximum bio absorption capacity qmax(mg/g) of Cr (VI) by various adsorbents (Farhan et al., 2012)

Bio sorbent	q_{max} (mg/g)	Reference
Hazelnut shell	17.7	Cimino et al., 2000
Almond shell	3.4	Pehlivan et al., 2010
Ground nut shell	5.88	Pehlivan et al., 2010
Gulohar fruit shell	12.28	Prasad et al., 2010
Hazel nut shell	8.28	Pehlivan et al., 2010
Walnut shell	8.01	Pehlivan et al., 2010
Walnut shell powder	138.89	Farhan et al., 2012

Table 2. compares the maximum biosorption capacity (qmax) of the powdered walnut shell with a few other biosorbents that have been documented in the literature. Variations in qmax are caused by the characteristics and nature of each biosorbent, including its surface area and major functional groups. The powdered walnut shell had a high Cr (VI) biosorption capability when compared to other adsorbents (Farhan et al., 2012)

Agriculture Application

Pesticide-residue removal

Biomass waste of walnut shells can also be used as an activated carbon to remove coal in soil better than the commercial activated carbon made of non-biomass. (Ahmad and Khuram Shahzad 2018) Walnut shell is popularly employed to remove soil residue that is abandoned by the pesticides. The Metolachlor (MET) can be eliminated in soil by the adsorption process with the help of Walnut shell biochar. It was modified by addition of original BC, kaolinite, illite, and montmorillonite. The rate at which the walnut shell and rice husk disintegrated was slow and therefore increased the degradation period of the bio composite. In thermogravimetric studies, it was established that an increase in the proportion of rice husk and walnut shell in biocomposites enhanced thermal stability in biocomposites. (Liu et al. 2022)

Biochar effect on plant

Biochar manufactured using different organic materials has been used to increase soil fertility and other soil characteristics and to boost seed germination. Also, biochar has been utilized to enhance the productivity of plants by enhancing cation exchange capacity to preserve the nutrient levels and lessening the impact of abiotic stressors such as drought by increasing soil water holding capacity (WHC) (Pluchon et al., 2014).

Moreover, augmentation of biochar to soil can enhance the carbon stocks within the soil and mitigate the adverse impacts of climate change by enhancing physicochemical characteristics of soil such as pH, cation exchange capacity, soil water holding capacity and plant-nutrient retention, microbial activity, and nutrient availability (Biederman et al., 2013). Besides, biochar use may enhance the aeration and structuring of soil through raising soil porosity and reducing the bulk density (Joseph et al., 2010). It helps in the rebuilding of the ruined soils and the maintenance of the nutrition of plants by absorbing and slowly releasing nutrients to plants (Sohi et al., 2010).

addition of biochar resulted in improvement of microbial activity in the soil and it produced an environment conducive to nitrification. He et al., The biochar has also been applied with high success as a sorbent (Sheng et al., 2005) by sticking to its surface that can reduce phytotoxic chemical levels as well as heavy metal concentrations that inhibit the development of plants and inhibit seed germination (Atkinson et al., 2010).

Food applications:
The shell of walnut is also a source of raw material that is renewable it remains unused after the kernels of walnuts had been used as food (Khantwal et al.,2016).

Oil extraction from walnut shell
In the walnut shell medium good oil adsorption capacity and low media gravity are obtained. They are strong enough to maintain their size during the backwashing operation so as to reduce the possible loss during the filtration process. The literature does not provide the oil sorption capacity and oil recovery of the walnut shell filters although the studies on the use of the filters as an alternative to the conventional sand filters have been conducted. Such research would be of value in comparing and selecting a sorbent to be used in a specific application. (Srinivasan et al., 2008)

Dye absorption:
The adsorption of heavy metals has been successfully made using good stability walnut shell powders (WSPs). Lignin and cellulose are considered some of the most prevalent amphiphilic compounds in the walnut shells. Lignin and cellulose contain a large amount of active groups (-OH and The use of (-OH and −COO−). WSPs to treat the water enables the achievement of such a type of waste utilization as agricultural residue. A multifunctional layer of water remediation using WSPs was developed in this work with the help of the crushed and purge process using the remains of walnut shells. This is simple and easy to apply unlike the complicated process that is involved in the production of conventional super hydrophobic or underwater super oleophobic filtering membrane. The presence of lignin and active groups (-OH and of cellulose(-OH and −COO−). and their higher capacity of absorbing water has made the WSPs layer to be poor in adhering oil and repelling oil underwater (Li et al., 2017).

Medical application
Dry eye disease
WS glasses have been applied in a study to offer therapeutic alternatives and evaluate the acceptability and effectiveness of moxibustion to manage patients with dry eye disease (DED). Moxibustion can be used with WS eyewear to normalize the tear film and clinical symptoms of patients with DED. Nonetheless it is not significantly more effective than sodium hyaluronate eye drops in terms of reducing the corneal damage and tear production (Zhang et al., 2022).

Anti-Cancer Activity
The green peel of the unripe fruit of Jregia, or walnut, has been used in traditional Chinese medicine with numerous therapeutic properties such as long-term treatment of malignant tumors. J. regia green peel contains flavonoids as the principal active ingredients and elements. Walnut shell, which is green, possesses anti-cancer effect (Alinia-Ahandani et al., 2019; Wei et al., 2022).

Therapeutic drug production

Proanthocyanides are phenolic chemicals present in nuts that are an indicator of potent antioxidants that are twenty times stronger than ascorbic acid. On its own, proanthocyanidins are a polymer chain of flavonoids, such as catechin. At this very stage, it is safe to say that the possibility of developing a waste-free processing model of the walnuts, identification of the groups of biologically active substances in the shell, and the justification of the economic viability of the studies are all relevant and encouraging directions in the science. These involve utilization of walnut shells as raw materials in the manufacture of drugs with diverse therapeutic and preventive properties depending on the release of remnant beneficial biologically active compounds. (Kizatova et al., 2022)

Conclusion

Edible shells of nuts are potential sources of biomass, and have the benefit of having a high concentration in the nut milling plant. Walnut shells, which belong to the family of Juglandaceae is an organic and biodegradable raw material that has great commercial advantages. The world produced more than 3,763.725 metric tons of walnuts in 2016 ,47 and 31 of which was produced in China and the United States, respectively. The ash obtained by burning the shell of walnut may be utilized as a catalyst, adsorbent or catalyst in the production of biodiesel. Activated carbon and carbon nanotubes can also be made using walnut shells and they have the ability to clean water. They find application in lignin-based adhesives, activated carbon, ceramic bricks that are fired, polymer composite, and the removal of pesticide related soil residue. Walnut shells may be moreover used in the pharmaceutical development with prophylactic and therapeutic action.

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