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Review Article

Rice Husk, Melon Husk and Shea Butter Husk as Potential Adsorbent for Removal of Metal Ions from Solution: A Review

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ABSTRACT

Activated carbon has been extensively used as adsorbent for the removal of heavy metals from effluent water generated by various industries. However, activated carbon remains an expensive material. Currently, the need for safe and economical methods for the removal of heavy metals has necessitated research towards the use of low-cost agricultural wastes for the production of adsorbents for treatment of heavy metal contaminated water and wastewater. In this review, production of activated carbon from low cost agricultural wastes such as rice husk, melon seed husk and shea butter husk is discussed.

1. INTRODUCTION

There are 1.1 billion people in the world who still do not have access to safe drinking water and the majority of them are in the developing and the world's poorest countries (Abaliwano *et al.,* 2008). Water pollution caused by human and industrial activities has been reported as a global problem (Satya *et al*., 2011). Water pollution is the universal cause of 80 % of diseases (Alfarra *et al.,* 2014).

Numerous industries (e.g., electroplating, metal finishing operations, electronic – circuit production, steel and non-ferrous processes and fine-chemical and pharmaceutical production) discharge a variety of toxic metals into the environment (Sankpal and Naikwade, 2012). Thousands of chemicals are discharged directly and indirectly into water bodies without treatment for the elimination of the included harmful compounds (Salim *et al.,* 2008). Heavy metals are without doubt the most hazardous and harmful metals even when present in traces, since they accumulate in the tissue of living organisms (Rao *et al.,* 2010). Table 1 shows the toxicity of some heavy metals and their sources.

Table 1: Heavy metals sources and its toxicity

Metal	Source	Toxic effect	Reference
Arsenic	Mining, smelting, energy production from fossil fuels and rock sediments	Haemolysis, liver tumors, gastrointestinal symptoms, cardiovascular, nervous system functions disturbances and bone marrow depression.	(Momodu and Anyakora, 2010)
Cadmium	Electroplating, smelting, alloy manufacturing, pigments, plastic	Hypertension, Itai–Itai disease, carcinogenic, lung insufficiency, bone lesions, cancers, weight loss	(Momodu and Anyakora, 2010; Sharma and Bhattacharyya, 2005; Singh and Hasan, 2005)
Chromium	Electroplating, paints and pigments, steel fabrication and canning industry	Vomiting, severe diarrhoea, lung tumors, Carcinogenic, pain, nausea	(Ali *et al.,* 2007; Rao *et al.,* 2010; Singh, 2005)
Copper	Paint manufacturing, wire drawing, copper polishing, and printing operations, Electronics plating	Dizziness, diarrhoea by nausea, vomiting, stomach irritation	(Ali *et al.,* 2007; Bilal *et al.,* 2013)
Lead	Electroplating, ammunition and Manufacturing of batteries	Brain damage, anorexia, malaise, loss of appetite anaemia and brain damage	(Ali *et al.,* 2007; Low *et al.,* 2000; Mataka *et al.,* 2006
Nickel	Electroplating, non-ferrous metal, mineral processing copper sulphate manufacture	Dermatitis, chronic asthma, chronic bronchitis and lung cancer	(Febrianto *et al.,* 2009; Öztürk, 2007)
Mercury	Volcanic eruptions, forest fires, battery	Corrosive to skin, eyes, muscles, neurological and renal disturbances	(Farooq *et al.,* 2010)
Zinc	Mining, manufacturing processes, smelting and ore processors	Gastrointestinal distress, nausea, diarrhea, fever, chills and chest pains	(Farooq *et al.,* 2010)

It is desirable to remove toxic heavy metals from wastewater before discharge into the water bodies. A number of synthetic methods have been reportedly used for the removal of metal ions from aqueous solutions. The methods are ion exchange, solvent extraction, reverse osmosis, electro dialysis, precipitation, flocculation and membrane separation processes (Blanes *et al*., 2016). However, these techniques have disadvantages that include high capital and operational costs, complex treatment and disposal methods for the residual metal sludge removal (Mohanty *et al*., 2006). Table 2 illustrates the advantages and disadvantages of some techniques used for the removal of heavy metals from solution.

Table 2: Advantages and disadvantages of some techniques for removal of heavy metals

Method	Advantages	Disadvantages
Chemical coagulation	Sludge settling Dewatering	High cost, large consumption of chemicals (Abaliwano *et al.,* 2008)
Chemical precipitation	Low-cost, easy. Most of the metals can be removed	Discarding problem. Huge amounts of sludge produced (Abaliwano *et al.,* 2008)
Electrochemical methods	No chemicals consumption Pure metals can be recovered Metal selective	High capital cost, high running cost (Rao *et al.,* 2010)
Ion-exchange	High regeneration of materials Metal selective	High cost. Smaller amounts of metal ions removed (Rao *et al.,* 2010)
Ultrafilteration and membrane process	Fewer chemical consumption High efficiency	High running cost Low flow rates Percentage removal of heavy metal decrease with the presence of other metals (Fu and Wang, 2011)
The use of natural zeolite	Most of the metals can be removed Reasonably less costly materials	Low efficiency (Fu and Wang, 2011)

Adsorption is a low cost and imperative physical process for the treatment of wastewater. In adsorption process metal ions are transferred from solution to the surface of the adsorbent and attached to the adsorbent (Bindra *et al*., 2013). Adsorption is a mass transfer process that involves the transfer of adsorbate from liquid phase into solid phase. It is segmented into 3 steps (i) Boundary layer mass transfer across the liquid film surrounding the surface of particle, (ii) Internal diffusion /mass transport within the surface particle boundary as pore and /or solid diffusion, (iii) Adsorption on the external surface (Gueu *et al*., 2007). Adsorption compared with other methods mentioned appears to be an attractive technique due to its efficiency and the ease with which it can be used for the removal of heavy metals from industrial wastewater (Rao *et al*, 2015). In recent years, a number of adsorptive materials, such as aquatic plants, agricultural by-products, industry by-product, sawdust, clay, zeolite and microorganisms were used in heavy metal removal from wastewaters (Bansal *et al*, 2009). Agricultural waste materials being economic and eco-friendly due to their unique chemical composition, availability in abundance, renewability and low cost seem to be a viable option for heavy metal remediation (Adebayo *et al*., 2016).

Several reports are available in the field of production of activated carbon from agricultural waste for the removal of heavy metals such as sugarcane bagasse, pith, nut shells, black gram husk, hazelnut, maize cob husk, walnut, almond, pistachio shell, and apricot stone (Igwe and Abia, 2006; Okafor and Aneke, 2006; Badmus *et al*., 2007; Onundi et al., 2010; Shafaghat *et al*., 2011).

Currently the production of activated carbon from agricultural waste has increased to a great extent. A general method for preparation of activated carbon from agricultural waste is shown in Figure 1.

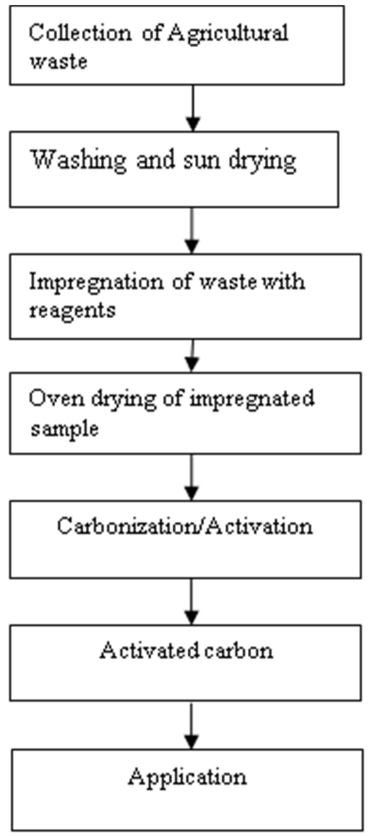

Figure 1: A general method of preparation of activated carbon from natural source

2. RICE HUSK, MELON SEED HUSK AND SHEA BUTTER HUSK AS ADSORBENT FOR HEAVY METAL REMOVAL

This review is focused on the use of rice husk, melon seed husk and shear butter husk as adsorbent for the treatment of heavy metal contaminated water. Rice husk essentially consists of 20% lignin, 25% hemicelluloses, 35% cellulose,17% ash (including silica) and 3% crude protein. This composition is associated with functional groups those make it suitable for metallic cations fixation (Chauhan, 2015), The surface of melon husk is covered with celluloses, lipids and proteins, hence adsorption process can occur on these organic functional groups (Adelagun *et al.*, 2014). While the husk from shea butter has high adsorption capacity, eco-friendly product and a low-cost adsorbent (Tsunatu *et al.*, 2015).

2.1. Rice Husk as Adsorbent

The third most imperative cereal crop grown around the world is rice, with a yearly production of more than 650 million tons. The major by-product derived from a rice processing mill is rice husk, which is a coat shielding the rice grain (Mohammad *et al.*, 2015). Rice husk consists of mineral ash (15.05%), water (8.11%), extractives (1.82%), cellulose (32.24%), lignin (21.44%), and hemicelluloses (21.34%). This composition is associated with many functional groups that make it suitable for metal ion removal (Mohammad *et al.*, 2015). Rice husk as an adsorbent in removing heavy metals has been reported.

Akhtar e*t al.* (2010) used activated carbon from rice husk activated by chemical and thermal activation with 0.1M HNO_3 and 1M K_2CO_3 at 473K. The obtained results showed that activated carbon produced from rice husk was used for removal of Pb(II), Cd(II), Zn(II) and Cu(II) from an aqueous solution over a pH range (1–10) via batch adsorption technique. The adsorption equilibrium was well defined by Freundlich, Langmuir and Dubinin–Radushkevish (D–R) isotherm models at equilibrium time of 20 min at pH 6 and using 0.2 g of sorbent. The kinetics of mass transfer and intra-particle diffusion for metal ions sorption onto RHA were studied with Lagergren and Morris–Weber kinetic models. The numerical values obtained from the thermodynamic parameters showed the exothermic nature, spontaneity and feasibility of the sorption process. Yahaya e*t al.* (2011), in a similar approach, investigated the use of activated carbon prepared from rice husk for adsorption of Cu (II) in batch mode with variation in operation parameters such as contact time, initial Cu(II) concentration and adsorbent dosage. The obtained result revealed that the low cost activated carbon prepared from rice husk was a good adsorbent for removal of Cu (II) from an aqueous solution. Langmuir and Freundlich isotherm models were used to test the adsorption data. Maximum monolayer adsorption capacity of Cu (II) onto RHA was 112.43 mg/g at 298 K. The pseudo-first-order and pseudo-second-order equations were used to study the adsorption kinetic. It was observed that the adsorption of Cu (II) onto RHA was best described by the Langmuir and pseudo-second-order respectively for adsorption equilibrium and kinetic studies. Hegazi (2013) has reported the use of activated carbon derived from rice husk for the removal of Fe(II), Pb(II), Ni(II), Cd(II) and Cu(II) from electroplating industry wastewater. The results obtained showed that the low cost adsorbent was successfully used for the removal of the heavy metal ions at a concentration range of 20–60 mg/l. The amount of removal was dependent on the adsorbent dosage and the adsorbate concentration. Elham e*t al.* (2010) utilized rice husk for the removal Zn(II) and Pb(II) ions present in dairy wastewater. The percentage adsorption of Zn(II) and Pb(II) ions increased with an increase in contact time and adsorbent doseage. The experimental data were analyzed by Langmuir isotherm. The maximum adsorption capacity of the adsorbent for Pb(II) and Zn(II) ions were found to be 0.6216 mg/g and 19.617mg/g respectively. Haris e*t al.* (2011) utilized unmodified rice husk (raw rice husk) and modified rice husk (RH) with NaOH, KOH, and $Ca(OH)_2$ for removal of Cd(II) ions in aqueous solution. Higher percentage removal was achieved using activated rice husk compared with the unmodified rice husk. The adsorption percentage increased in the order of unmodified RH (80.13 ±0.46%) < $Ca(OH)_2$ –modified RH (90.74 ± 0.18%) < NaOH-modified RH (93.36 ± 0.23%) ≤ KOH-modified RH (93.78 ± 0.27%).

2.2. Melon Seed Husk as Adsorbent

Husk from melon seed (*Citullus colocynthis*) are readily available and are good raw materials for adsorbent for removal of heavy metals like cadmium and lead (Daniel *et al.*, 2014). Melon (*Citrullus colocynthis*) belongs to the class of the genus *Citrullus* of *cucurbitaceae* family. Curcubits are well-known for their high oil and protein content. Seeds from cucurbits are sources of protein and oils with up to 35 % protein and about 50% oil (Solomon *et al.*, 2010).

Results of pervious work conducted on development of adsorbent from melon seed husk indicates that melon seed husk is an attractive and inexpensive alternative for the removal of heavy metals from water and wastewater (Daniel and Chinedu, 2014). Daniel and Chinedu, (2014) used melon husk activated with urea (activated melon husk) as adsorbent to remove cadmium from industrial wastewater. Laboratory experiments were carried out to identify the effect of adsorbent dosage, contact time, and effect of initial metal concentration. Result of their findings showed that, percentage removal of cadmium increased with increasing carbon dosage, with maximum removal attained with 0.8 and 1.0g. Maximum removal was obtained at a contact time of 20 minutes with 97.6% adsorption. Application of adsorption isotherms had positive correlation with both Langmuir and Freundlich isotherm models. In a similar manner, Daniel *et al.* (2014) used melon seed husks activated with sulphuric acid (H_2SO_4), to remove cadmium and lead from industrial effluents. Equilibrium isotherms, kinetics study and adsorption studies of process variables, such as contact time, metal ions concentration and adsorbent dosage were studied in batch experiments. Cadmium removal was found to be dependent on the three parameters with maximum removal attained in 70 min with

0.6g activated melon husk and also maximum adsorption occurred at minimum concentrations of metal ions. Adsorption isotherms correlated well with both Langmuir and Freundlich isotherm models and their R^2 values range from 0.911- 1. Adsorption process for the two heavy metal ions was found to follow pseudo-second order adsorption kinetics. Adelagun *et al.* (2014) in another study investigated the use of modified melon seed husk (MMH) in remediation of water contaminated with lead ions. The modification was done by soaking the melon seed husk in 0.5M NaOH solution. The influences of pH (2-6), metal ion concentration (25mg/l – 100mg/l) and adsorbent dosage (0.05g - 0.60g) were studied. The equilibrium sorption isotherm was studied using Freundlich and Langmuir isotherm models. The results obtained from the model showed that the process of the sorption fitted better with the Freundlich isotherm, implying that sorption occurred on a heterogeneous surface of the MMH. Babarinde *et al.* (2014) reported the uptake of Co(II), Ni(II) and Cr(III) from aqueous solution onto melon seed husk (MSH). The biosorption of each metal ion was dependent on the solution pH, biosorbent dose, initial metal ion concentration, contact time and temperature. Isotherms and kinetic models were applied to the biosorption process in order to establish the best models for the biosorption of these metal ions from solution. The Freundlich isotherm model gave the best fit with the highest correlation coefficient (R^2). The kinetic study showed that the biosorption of Co(II) Ni(II) andCr(III) onto MSH best followed pseudo-second-order kinetics. The thermodynamic parameters showed that each of the processes was endothermic, the order of spontaneity was found to be Ni(II)>Co(II)>Cr(III), while the order of disorder was found to be Ni(II)>Cr(III)>Co(II).

2.3. Shea Husk as Adsorbent

Shea crop grows extensively in sub-Saharan Africa and is in high demand from numerous world markets. ln West Africa, an estimated 600,000 tons of shea nuts are harvested and about 350,000 tons are exported (Tsunatu *et al.*, 2015). In Nigeria, an estimated 250,000 metric tonnes of shea nuts are harvested per annum (Yahya *et al.*, 2013). The husk from shea butter has high adsorption capacity, is an eco-friendly product and a low cost adsorbent (Tsunatu *et al.*, 2015).

Tsunatu *et al.* (2015) studied the adsorption of cyanide ions by shea butter seed husk (SBSH). The experiments were carried out in batch reactors with best operating parameters at 100 mg/l for the initial cyanide concentration, pH value 10, temperature 30±2ºC, adsorbent dosage (3.0mg/100 ml) and contact time of 120 minutes with maximum percentage cyanide ion removal of 94.56%. The values of cyanide ions adsorbed at equilibrium (qe) and amount adsorbed at a given time (qt) of the experimental results were fitted to Freundlich, Langmuir and Temkin isotherm models with Temkin adsorption isotherm having R^2=0.966, and best describing the equilibrium model. Yahya *et al.* (2013) in another study investigated the use of modified shea butter cake for the removal of Cu^{2+} ions from aqueous solution. The shea butter cake was modified with calcium alginate. Instrumental analysis such as Scanning electron microscopy (SEM), X-ray Diffractometer (XRD) and Fourier-transform infrared spectroscopy (FT-IR), were used to characterize of the solid sorbent. Batch equilibrium study was carried out to evaluate the adsorption capacity, and process parameters such as contact time, pH, initial metal ion concentration, and adsorbent dosage were studied. An optimum pH of 5, equilibrium time of 30mins and adsorbent dosage of 40 beads was obtained. The Langmuir and Freundlich isotherms were used to fit the experimental data and values fitted well into the Langmuir more adequately with correlation coefficient unity 1.000 at 10mg/l of initial metal ion concentration. The kinetic study using the pseudo-first-order and pseudo-second were used to determine the rate constants. The experimental data fitted well into pseudo-second–order better. The results indicated that modified shea butter cake could be employed as an adsorbent for the removal of copper (II) ions in an aqueous solution.

3. CONCLUSION

There is increasing interest in the preparation of low-cost adsorbent from agricultural waste as an alternative to commercial activated carbon for both wastewater and water treatment. Presently, rice husk, melon seed husk and shea butter husk have shown potential to be used as adsorbent for wastewater treatment in a cost-effective way. The use of agricultural wastes helps to overcome part of the excessive agricultural wastes in the environment. The use of commercially available activated carbon for the removal of the heavy metals can be replaced by the utilization of inexpensive, effective, and readily available agricultural by-products as adsorbents.

4. CONFLICT OF INTEREST

There is no conflict of interest associated with this work.

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