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An emerging adsorption technology and its applicability on trees as an adsorbent for the remediation of water pollution: A review

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ABSTRACTS

Wastewater pollution and its treatment is an ever-increasing concern in the century because of the higher industrialization and urbanization. The expansion of industries is the leading cause of polluting water resources. A discharge from industries contains various organic and inorganic pollutants. The heavy metals which are mostly discharged from the industries include Zinc (Zn), Arsenic (As), Nickel (Ni), Cadmium (Cd), Mercury (Hg), Copper (Co), Chromium (Cr), and Lead (Pb). These metals are toxic to human health and other living organisms. Water is an essential natural resource that reserves important flora and fauna on the earth. Therefore, it is necessary to remediate contaminated water from organic and inorganic pollutants. Chemical precipitation, ion exchange, reverse osmosis, chemical oxidation, reduction ultrafiltration, electrolysis, and adsorption are some treatment processes used to remove heavy metals from wastewater effluent. Adsorption is the most promising technique used for the removal of water pollutants. This review surveys the various natural adsorbents used to remove water pollutants. Natural adsorbents are the most effective and low-cost adsorption techniques in which plant residuals (leaves, stems, roots, straw, etc.) are used to remove water pollutants from wastewater.

Key words : *Adsorption technology, Remediation wastewater, Removal of heavy metals, Water pollution, Natural adsorbent, Tree adsorbent.*

Introduction

The most common contaminants in water areas are heavy metals, mainly contributed by anthropogenic sources like fabrication, battery manufacturing, paper, and pulp industries, iron industries, fertilizer and pesticides industries, electronic assembly, textile industries, and mining activities (Obuseng *et al.*, 2012). Water pollution is the leading cause of 80 % of diseases (Alfarra *et al.*, 2014). Water pollutants are heavy metals generally considered those whose density exceeds 5 g per cubic centimeter. Heavy metals are considered as the following elements: Aluminum (Al), Manganese (Mn), Copper (Cu), Cobalt (Co), Molybdenum (Mo), Nickel (Ni), Arsenic (As), Selenium (Se), Chromium (Cr), Silver (Hg), Zinc

(Zn), Iron (Fe), Cadmium (Cd), Tin (Sn), Lead (Pb), and Gold (Gunatilake, 2015). Their presence in the water bodies causes severe threats to flora and fauna dependent on the water bodies (Babel and Kurniawan, 2004). They can be absorbed and accumulated in the human body through food and drinking water and have serious health effects like cancer, organ damage, nervous system damage, and in extreme cases, death (Gunatilake, 2015). Some toxic metals likes Cu, Ni, and Zn are higher than industries permissible discharge levels in these effluents (Parmar and Thakur, 2013). According to the US Environmental Protection Agency (EPA), Pb, Cu, Cd, Mn, Zn, and Ni in drinking water must not exceed 0.015, 1.3, 0.005, 0.05, 5and 0.04 mg l^{-1} (Sheng *et al.,* 2004;) Therefore, it becomes necessary to remove these water pollutants from this wastewater with appropriate treatment before releasing them into the environment. There are various methods to remediate of water pollutants from wastewater, i.e., ion exchange, chemical precipitation, reverse osmosis, chemical oxidation, reduction ultrafiltration, electrolysis, and adsorption. These are some treatment processes used to remove heavy metals from wastewater (Fu and Wang, 2011). Among these methods, adsorption is the most promising technique to remove water pollutants. Biosorbents, which are found in nature in the form of bio-waste mainly plant and tree waste, and have the capacity to remove water pollutants from wastewater. Adsorption provides a number of benefits such as traditional treatment procedures, the potential to reuse materials, and low in cost (Demirbas, 2008). This paper will discuss the adsorption technique and how tree parts are used as an adsorbent for the removal of heavy metals from polluted water.

Biosorption is a property of living and non-living organisms (Bilal *et al*., 2013). It refers to removing biological materials from the solution (Gadd, 2001). An interaction between a biosorbent (a solid surface consisting of a biological matrix) and sorbate (atoms and molecules) is known as "biosorption," and it results in a deposition of the sorbate on the sorbent surface so, a decrease in the concentration of sorbate. The process of removing metal ions from aqueous solutions usually uses the dead waste material of biomass (Sasaki *et al*., 2013). The Significant advantages of biosorption technology include low-cost, high efficiency, no additional nutrient requirement, and regeneration of the biosorbents (Fu and Wang, 2011; Ahalya *et al*., 2003. Adsorption studied mainly focused on untreated plant wastes such as grape stalk wastes pellets of peanut hull (Watson, *et al*., 2002), neem bark, rice husk ash (Mandal *et al*., 2006), 1 *Moringa oliefera* pods (Adelaja *et al*., 2011), tea waste (Kailas, 2010), and sago waste (Wase *et al*., 1998).

Composition of plant waste

Plants usually comprise lignin and cellulose as the main constituents (Beveride and Murray, 1980). Other elements are hemicelluloses, lipids, ash, water, hydrocarbons, simple sugars, carbohydrates, proteins, and water, etc. these are found in the plant and contain a range of functional groups involved in the binding process (Gupta and Ali, 2000). These groups are assumed to bind heavy metal by replacing hydrogen ions for metal ions in solution or by giving an electron pair to form complexes with the metal ion in the solution (Alfarra *et al.,* 2014).

Preparation of adsorbent

Collection of tree samples leaves (Sharma and Bhattacharyya, 2005; Hanafiah *et al*., 2006; Rao *et al*., 2010), stems (Tan and Xiao, 2009; Sun and Shi, 1998; biomass (Sarada *et al*., 2017), bark (Al-Asheh and Duvnjak, 1997; 2007; Seki *et al*., 1997) and seeds (Flores-Garnica *et al*., 2013; Obuseng *et al*., 2012), etc. Samples were washed several times with tap water to remove all the dirt and other extraneous depositions. The materials were rewashed thoroughly with double distilled water. The material was sun-dried for ten days, then in a microwave for about 30 min (40 – 100 ^{0}C), and then dried material was a grind and sieved into uniformed size particles. Powdered material was kept in an airtight container when required Gopal *et al*., 2014).

Biosorption process

Batch adsorption studies were carried out by adding a known weight of prepared adsorbent (extract powder from leaves, stems, biomass, barks, seeds, etc. to the metal ion solution of 200 ml under different test conditions. The effect of solution pH, contact time, initial concentration, and doses of leaf extract powder with diverse mixtures and temperatures (Dubey *et al.,* 2013; Wang *et al.,* 2010; Celebi, *et al*., 2020) date were recorded. All the adsorption experiments were carried out at room temperature (25 ± 2 ^{0}C). The pH will adjust to 2-5, every 20 minutes with 1M HNO_3 solution (Zvinowanda *et al*., 2009). The amounts of metal adsorbed by the biosorbent per unit mass were calculated as:

$$q = \frac{(Co - C)V}{m}$$

Where, Co = the initial metal concentration, C = the equilibrium metal concentration (mmol-L^{-1}), *m*- the mass of the adsorbent (in grammes), and *V*- the volume of the solution (ml), and q = the metal adsorbed at equilibrium (mmol-g^{-1}). Percentage of metal ion removed (%) was calculated using the equation (Desta, 2013):

$$q\,\% = \frac{(Co - C)}{Co} X100$$

Adsorption mechanism

Adsorption is an exothermic surface-based process in which molecules of a gaseous or liquid substance accumulate on an adsorbent surface (Singh *et al.*, 1998). The adsorbate is the substance that is adsorbed on the adsorbent. While the opposite process of adsorption is known as desorption. Figure 1 shows a schematic representation of this procedure.

There are two ways of adsorption process based on the adhesive of molecules on the surface of the adsorbent, which is "physical adsorption" (also called physisorption) and "Chemical adsorption" (also called chemisorption). Physisorption is the weaker force i.e., The Van der Waals forces and electrostatic attraction, and is formed of thick multilayer adsorption on the surface while chemisorption is the strong chemical bond, i.e., covalent bond, and usually develops monolayer adsorption (Dabrowski, 2001). This process is illustrated in gure 2 A and B.

Adsorption isotherm models

Equilibrium modeling

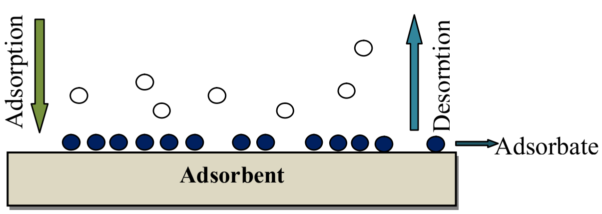

Fig. 1. Schematic representation of this procedure (Kecili and Hussain, 2018).

The highest adsorption of metals at different changeable parameters in the biosorption process is known as an equilibrium model. The isotherm model is the relationship between the amount of metal adsorbed by the adsorbent and the remaining metal concentration in the solution (*et al.*, 2016). The equilibrium isotherm can be expressed through Temkin, Dubinin-Rudushkevich (D-R), Freundlich, Braunure-Emmett Teller (BET), Langmuir models. Freundlich and Langmuir model is the most frequently used isotherm equilibrium model, and it is applicable to batch absorber systems (Demirbas, 2008;)

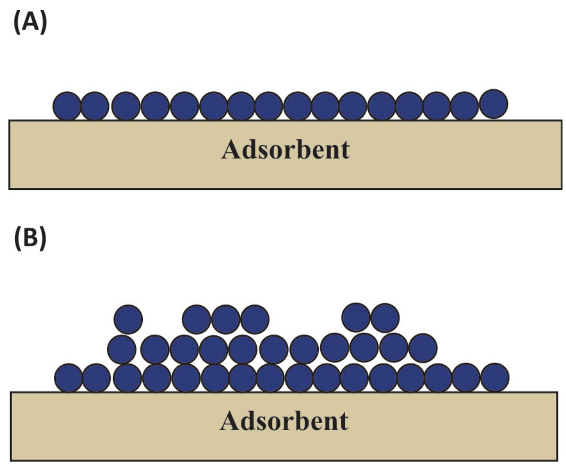

Fig. 2. Monolayer adsorption A and multilayer adsorption B

Langmuir isotherm

This model was proposed by Irving Langmuir (Langmuir, 1918) and the main assumptions of the isotherm model are (Vilardi *et al.*, 2017; Jian *et al.*, 2016;)

- It refers of the adsorption upon homogenous area.
- Monolayer adsorption takes place on the surface of the adsorbate
- The adsorption only occurs at a finite (fixed) number of definite localized sites.

The given equation can demonstrate the Langmuir isotherm:

$$q_e = \frac{q_{max} b c_e}{1 + b c_e} \quad \text{.. (i)}$$

Here,

q_e - The quantity of metal sort at equilibrium (mg/g),

q_{max} - The sorption capability of monolayer (mg/g),

b- Adsorption equilibrium constant (L/gm),

c_e - The amount of metal ions present in an aqueous solution (mg/L) (Jian *et al.*, 2016 Kecili and Hussain, 2018).

Freundlich isotherm

The Freundlich isotherm model proposed by Herbert Freundlich (Freundlich, 1906) and main assumption of the isotherm are:

- The adsorption process occurs on multilayer of absorbent molecules.
- Heterogeneous adsorption occurs on the molecule of the adsorbent surface.

The equation can demonstrate this isotherm:

$$\log q_e = \log K_F + \frac{1}{n} \log C_e \quad .. \text{(ii)}$$

Here, - the amount of absorbed (mg/g), - the equilibrium value of adsorbate ions (mg/l), (l/g) and *n* is Freundlich constant and Freundlich exponent, respectively. (mg/g) indicates the adsorption capacity of the adsorbent toward the adsorbate, and *n* is an indicator for the degree of the surface heterogeneity and describes the distribution of the adsorbate molecules on the adsorbent surface. A value of *n* higher than 1 indicates favorable molecules' adsorption onto adsorbent surfaces (Jain *et al*., 2016; Kecili and Hussain, 2018).

Temkin isotherm

Temkin and Pyzhev proposed this isotherm model (Temkin and Pyzhev, 1940a; Temkin and Pyzhev 1940b). This model considers the effect of adsorption heat, which linearly decreases with the adsorbate molecule layer's coverage. The adsorption heat decreases due to the interaction between the adsorbed molecules on the surface (Kecili and Hussain, 2018).

The following equation illustrates the Temkin adsorption isotherm:

$$q_e = \left(\frac{RT}{b_T}\right) \text{In } (AC_e \quad \text{.......... (iii)}$$

Where *T* - the temperature (K), *R*- the ideal gas constant (8.314 $Jmol^{-1}K^{-1}$), represents the Temkin constant ($Jmol^{-1}$), which depends on the adsorption heat, and *A* is the equilibrium adsorption constant, which corresponds to the maximum adsorption energy (L mg^{-1}).

Brunauer-Emmett Teller (BET) isotherm model

This isotherm theory was given by Stephen Brunauer, Paul Hugh Emmett, and Edward Teller in 1938 there; for is known as Brunauer-Emmett-Teller (BET) theory. It is similar to the Langmuir isotherm theory.

- It represents the multilayer adsorption of the molecules to the adsorbent surface.
- This isotherm used for the adsorption process in gas solid systems.
- It is used to measure the surface area of solid or porous materials.
- BET equations describe the relationship between the numbers of gas molecules adsorbed at a given relative pressure.
- Adsorption at one site does not affect adsorption at another site.

The following equation illustrates the Brunauer-Emmett Teller (BET) adsorption isotherm:

$$\frac{1}{v\left[\left(\frac{p0}{p}\right)-1\right]} = \frac{c-1}{v_m c}\left(\frac{p0}{p}\right) + \frac{1}{v_m c} \quad .. \text{(iv)}$$

Here, *p* – equilibrium, *p0*- saturation pressure at the adsorption temperature, *v* - adsorbed gas quantity, – quantity of adsorbed in monolayer sorption, and *c* is the BET constant (v).

$$c = \exp\left(\frac{E_i - E_L}{RT}\right) \quad .. \text{(v)}$$

E_1 - heat of the first layer of adsorption, and E_L is that for the second and higher layers and is equal to the heat of liquefaction or heat of vaporization.

Dubinin- Rudushkevich (D-R) isotherm

This model was proposed by Dubinin – Astakhov, and Dubinin – Radushkevich and the following assumption of the isotherm:

- Molecular adsorption occurs in the microspores instead of adsorption on the adsorption surface, which leads to the monolayer of multilayer formation.
- It is commonly applied to the adsorption processes of the subcritical vapors in the pores of the adsorbents.

The equation demonstrates this isotherm model:

$$\text{In} \quad q_e = \ln q_m - \beta E^2 \quad .. \text{(vi)}$$

Where,

q_e = The amount of adsorbate molecules per mass of adsorbent (mg g^{-1}),

$q_,$ = Represented the absorption capacity (mg g^{-1}),

β = the activity coefficient (mol^2 J^{-2}), which represent the adsorption energy, and

E=Demonstrates the Polanyi potential as given in the following formula:

$$E = RT\text{In } 1 + \left(\frac{1}{C_e}\right) \quad .. \text{(vii)}$$

Here, R - ideal gas constant (J mol^{-1} K^{-1}), T - temperature (K), and C_e = the concentration of the molecules at equilibrium (mg L^{-1}).

Factor affecting biosorption

Following factors are affecting the biosorption process:

Temperature: The biosorption process exhibits either exothermic or endothermic thermodynamic behaviour. Positive change value indicates an exothermic reaction, which makes lower temperatures acceptable for metal ion sequestration. The negative enthalpy change represents an endothermic reaction, i.e., The degree of metal ion adsorption increases with temperature (Jain *et al.*, 2016). It seems not to influence the biosorption parameters in the range of 20-35 ^{0}C (Aksu *et al.*, 1992), but it mostly depends upon the features of the heavy metal. Ismail *et al.* (2009) investigated the batch experiment temperature impacts of powdered maize cobs for cadmium ion removal. The greater adsorption capacity for cadmium ions was observed 18.15 mg/g at 250 °C and 25.51 mg/g at 550 °C. The metal uptake increase with the decrease of temperature from 40 ^{0}C to 10 ^{0}C (Aslam *et al.*, 2010).

pH: It impacts the chemistry of metal solution, the reactivity of biomass functional group, and metallic ion competition. (Ferris and Myers-Keith, 1992; Galun, 1987). Wang *et al*. (2010) investigated on pH effect of Cr (VI) biosorption capacity of wheat residue. The studies recorded the maximum biosorption capacity received at 1.0 pH. Another study observed by Chen *et al.* (2010) showed an initial effect of pH on the biosorption capacity of Cu (II) used *Cinnamomum camphora* leaves and reported that raising pH value enhanced Cu (II) adsorption ability. Biosorption capacity increased with rising pH values (Flores-Garnica *et al.*, 2013; Sharma and Bhattacharya (2005). Dharmambal *et al*. (2015) studied on adsorption of Rhodamine –B dyes in the aqueous solution by *Tectona grandis* bark powder. The maximu m adsorption was found at 81.93% (2-8 pH).

Initial concentration of metal: At greater concentrations, the biosorption capacity increases and higher concentration provides a powerful pulling force to take over the metal's impediment to energy transfer between the solid and aqueous phases. It depends upon the properties of heavy metals, which are removed by the adsorbent (Yang *et al*., 2016; Ghasemi *et al*., 2017; Celebi *et al*., 2020). Lead (II) absorption of bark of *Moringa oleifera* increased at increases concentration (Reddy *et al.* 2011). Investigated on shells of oil palm for expulsion of Copper (II) and Lead (II), and the result was shown that the adsorption of Cu (II) and Pb (II) ion was increased with the increase of initial concentration of the solution.

Available heavy metals into solution: Biosorption is mainly used to treat wastewater containing various metal ions. However, the group of metal ions present in the source may affect the absorption of one metal ion (Sakaguchi and Nakajima, 1991).

Dosage of adsorbent: Adsorption of metals increases with higher adsorbent dosages because the maximal adsorbent provides the surface area or binding site. Celebi *et al.* (2020) looked at Brewed tea waste (BTW) for the adsorption process and found that the increases in BTW doses from 0.1 to 5 g in 100 ml, with an increase in the removal efficiency for Pb from 49.71% to 98.03%, for Ni from 36% to 76%, for Zn from 46.61% to 85.03% and Cd from 41% to 94%, respectively. The effect of adsorbent dose on the adsorption process of heavy metal onto the Activated Teff Straw (ATS) was investigated. The metal adsorption efficiency increased with the adsorbent dose (Desta, 2013).

Agitation time: Adsorption capacity increased with increase the agitation time. Telkapalliwar and Shivshankar (2017) investigated *Acacia nilotica* bark as a suitable an inexpensive adsorbent for removing Zn (II) ions from water through batch adsorption. The removal of Zn (II) ions increased slowly with time up to 60 minutes and then grew slowly. With increasing contact time, the percentage of Pb (II) ions removed steadily increased, and equilibrium was attained in around 360 minutes (Waghmare and Chaudhary, 2013).

Particle size of adsorbent: The smaller particle size allows for even more reactive high adsorption sites for metal ions on the adsorbent's surface. Ozdes *et al.* (2014) studied the bark particle size affected the biosorption of Cr (VI) ions. Different sizes of the bark powder (like 150, 150-335, and 350-700 ìm) were selected for adsorption. Metal ion biosorption increased as the particle size of the bark decreased (Chong *et al.,* 2013).

Advantages and limitations of tree adsorbent

In nature, trees or plant-based adsorbents are usually inexpensive, freely available, and low operational cost (Gupta and Suhas, 2009).

- Adsorbents to be reused through the desorption process (Dhir, 2014).
- The adsorption technique can deal with various heavy metals in mixed wastes.
- In many situations, high affinity reduces residual metals to below 1 ppb.
- Metal adsorption may be accomplished in a relatively short contact period.

The drawbacks of using tree-based adsorbents for elimination of heavy metals are listed as follows:

- It requires modifying the form of the adsorbent, which may increase the treatment cost.
- An additional expense is incurred by gathering tree adsorbents and transporting them to a processing site.
- Some bioadsorbents are suitable for selective metal ions (Ahmad *et al*., 2012).

Tree-based adsorbent used for removal of heavy metals

Leaves as an adsorbent

Sharma and Bhattacharya (2005) investigated on neem leaf powder (NLP) as a biosorbent for the removal of Cd (II). They recorded that the adsorption was increased from 8.8% at pH 4 to 70% at pH 7.0 and 93.6% at pH 9.5; maximum adsorption was observed within 300 minutes of agitation. The Neem (*Azadirachta indica*) leaves powder prepared and tested on the removal of Lead (II) and adsorbent were considerably high Langmuir monolayer capacity of 300mg/g. Results were observed a small amount of adsorbent (1.2 g/L) was removed 93% of Pb (II) in 300 minutes from a solution of concentration of 100 mg/l at 300K (Bhattacharya and Sharma (2004). Rao *et al.* (2011) investigated of adsorption effectiveness of Cadmium (II) on *Ficus religiosa* leaf powder and observed that the Cd (II) adsorption was increased from 1.38% at 2 pH to 75.17% at 4 pH and 77.52% at 5.5 pH. Rao *et al.* (2010) investigated of adsorption capacity of Cd (II) ions on *Tectona grandis* leaf powder (TLP) and reported that 1 gm of TLP was removed 86.73% of Cd (II) from 50 ml aqueous solution containing 100mg/L of Cd (II) in 30 minutes of agitation. *Terminalia catalpa* leaves powder was used as an adsorbent to remove Cd (II), and adsorption percentages were increased from 0.86 mg/L at 2pH to 13.79 mg/L at 4 pH and 14.12 mg/L at pH 5.5 (Rao *et al.*, 2010). Similarly, Bhattacharya and Sharma (2004) studied *Azadirachta Indica* leaf powder and investigated the effectiveness of adsorbents for removing dyes from an aqueous Congo red solution. Results were recorded that 0.6 g $^{-1}$ of the Neem leaf powder could remove 52.0-99.0% of the dye from an aqueous solution with the agitation time increasing from 60 to 300 minutes. Bhattacharya *et al*. (2009) studied *Azadirachta Indica* leaf powder to remove Ni (II) from solution, and data were observed in various conditions, i.e., pH, adsorbent doses, temperature, and interaction time. The adsorption of Ni (II) ion increased according to the pH range between 2.0 to 5.0, with approximately 92.6% adsorption at pH 5.0 for the highest amount of the adsorbent (4 g/L). Dhabab *et al*. (2012) studied the *Ceratophyllum demersum* modified leaf powder to remove Cadmium ions and observed that 1.5g adsorbent was removed 95% cadmium ions at the 6.0 pH with 60 minutes of contact time. *Psidium guajava* leaves powder was prepared for adsorption of Cadmium ions, and the percentage of cadmium ions removal was increased from 74.0 % to 95.11 %, with an increase in pH range from 1.0 to 4.0 (Verma *et al*., (2010). Muniraj *et al*. (2019) investigated on *Citrus lemon* leaves powder for the removal of heavy metals effluent by textile industries, and data were observed that 80-90% of toxic metals removed 2.0 g of *Citrus lemon* leaves powder (in 50 ml of effluent). Prepared adsorbent of *Cinnamomum camphor* leaf powder for the removal of copper ions from an aqueous solution and adsorption of the Cu (II) tended to increase with increasing pH value from 2 to 3 (Chen *et al*., 2010). The adsorptions of Cd (II) ions from aqueous solution by *Hevea brasaliensis* leaves powder (HBLP) were investigated. The Langmuir model determined the higher adsorptions about 3.68 mg/g (Hanafiah *et al*., 2006). Reddy *et al*. (2010) prepared *Moringa oleifera* leaves powder for adsorbent to removal of Lead (II) from an aqueous solution. Langmuir model provided the best correlation with biosorption capacity of 209.54 mg g^{-1} at 313K. The presence of common metal ions like Na^+ K^+, Ca^{2+}, and Mg^{2+} does not affect Pb's adsorption (II). Hamdaoui (2009) and Olive tree (*Olea europaea*) were proposed as a novel low-cost non-conventional sorbent for removing cadmium from aqueous solution, and the highest adsorption capacity was observed at 64.94 g g^{-1}. Rao (2010) investigate on the *Syzygium cumini* leaves powders (SCLP) for removal of Cd (II). The maximum adsorption capacity of *Syzygium cumini* leaves powder at room temperature was estimated to be 34.54 gm g^{-1}. Qaiser *et al.* (2008) studied *Ficus religiosa* leaves powder for adsorption of lead ions and recorded the maximum biosorption capacity of lead at 37.45 mg g^{-1}.

Bark as an adsorbent

Telkapalliwar and Shivshankar (2017) investigated on *Acacia nilotica* bark as an inexpensive adsorbent that was suited for removing Zn (II) ions from aqueous solution. The maximum Zn (II) removal percentage was about 92.40 % at pH 4.0. The potential to remove Pb (II) from aqueous solutions through adsorption was used *Ailanthus excelsa* tree bark, and the data were recorded removal of Pb (II) ions at 53.5 % at pH 6.0 with 360 minutes of contact time (Waghmare and Chaudhary, 2013). The low-cost adsorbent of Cajeput bark was prepared for the removal of methylene blue from an aqueous solution, and results showed that the percentages of removal of methylene blue increased from 83.12 % to 98.35 %, with increased pH of the solution from 2 to 7 (Sukpreabprom *et al.*, 2021). John *et al.* (2011) studied the possible use of *Cassia siamea* to remove Cd (II) ions from an aqueous solution. The highest Cd (II) ions removals were achieved at 9.81 mg/g at pH 7.0. Saeed *et al.* (2020) studied the Biosorption of hexavalent chromium metal ions from an aqueous solution by the bark of *Cinnamomum verum.* The maximum adsorptions were achieved at pH 1.6 to 2.7 and adsorption 99.42 % to 99.51, respectively. The adsorption process of metal ions (Cr^{3+}, Cu^{2+}, Ni^{2+}, Pb^{2+}, and Zn^{2+}) from the aqueous solution used in coniferous barks was investigated. The greatest binding capacity of barks was recorded, Cr^{3+} is followed by the descending sequence of Cu^{2+}, Pb^{2+}, Ni^{2+}, and Zn^{2+} (Martin-Dupont *et al.,* 2002). Saliba *et al.* (2001) studied on eucalyptus barks for the adsorption of heavy metal ions. The adsorption capacities of heavy metals were recorded to be 2.61, 0.71, 2.24, and 0.75 mmol/g adsorptions for Cu^{2+}, Cr^{3+}, Cd^{3+} and Ni^{2+}, respectively. Biosorption of Ni (II) ions from electroplating wastewater by modified *Eribotrya japonica* (loquat bark) and the maximum adsorption capacity were observed at 27.548 mg/g (Salem and Awwad, 2011). Adsorption of zinc (II) by the modified *Eucalyptus sheathiana* bark was investigated. The process was firmly pH dependent, and the adsorption percentage of Zn^{2+} decreased with the increase in solution pH from 2.5 to 5.1. Similarly, the adsorption percentages of Zn^{2+} were reduced with the increase in adsorbent dosage, initial metal concentration, temperature, and ionic strength (Afroze *et al.,* 2016). Utility of *Eucalyptus tereticornis* bark for the remediation of acid mine drainage. Pretreatment of the acid mine water with tree bark followed by bio-removal using *Desulfotomaculum nigificans* resulted in about 75% and 84% respectively of sulphate reduction at pH 4.1 and 5.5 (Chockalingam and Subramanian, 2009). Aoyama and Tsuda (2001) investigated on the *Larix leptopels* tree bark as a biosorbent for removal of Cr (VI), and the maximum adsorption was recorded at 99% at pH 3.0 with 72 hours of agitation time. Egwuonwu (2013) studied on Neem, Mango, and Locust bean bark for adsorption of methyl red and methyl orange. The percentage removal of methyl red and methyl orange was 11% at 6.5, 4 pH, respectively. In comparison, mango tree bark powder was removed 15% at 2 and 6 pH, respectively, and Locust bean tree bark was removed 12% at 9 and 4 pH, respectively. Reddy *et al.* (2011) studied on the *Moringa oleifera* as an adsorbent for the removal of Nickel (II) from an aqueous solution. Ni (II) sorption was increased with the increased pH from 3.0 to 6.0. There was a gradual decrease in NI (II) biosorption adsorption with an increase in the number of cycles from 98.02% to 92.02% after a sequence of six cycles. Shrivastava and Rupainwar (2011) conducted a comparative evaluation for dye adsorption on neem and mango bark powder. The percentage removal of dye by neem and mango bark powder was observed at 5.0 and 2.0 pH, respectively. Argun *et al.* (2010) investigated Nickel adsorption on the modified pine tree bark. The maximum removal efficiency level obtained is 97% for modified pine bark at 8 pH and 80% for modified pine cone at 8.0 pH. Adsorptions of lead in industrial effluent by *Pongamia pinnata* bark powder were investigated. The maximum adsorption was found at 34.36 mg/g in effluent at pH 5.0 - 6.0 and temperature of 30^{o} C for 60 to 65 minutes of agitation (Mamatha *et al.,* 2012). Similarly, Dharmambal *et al.* (2015) reported the adsorption of Rhodamine –B dye in the aqueous solution by *Tectona grandis* bark powder. The maximum adsorptions were observed at 81.93% at 2-8 pH for 50-60 minutes of agitation time.

Seeds or pods, or fruits used as an adsorbent

Festus *et al.* (2013) investigated on the flame of the forest (*Delonix regia*) tree pods for the sorption of lead and nickel ions from solution. The result showed that *Delonix regia* effectively removed these elements from aqueous solution, and equilibrium sorption of both metals was attained within 60 minutes of interaction and 98% removed metals. Qahtani (2015) reported water purification using banana, kiwi, and tangerine fruit waste by removing heavy elements Cd^{2+}, Cr^{3+} and Zn^{2+} ions. The order of maximum adsorption capacity of metal ions on banana was recorded $Cr^{3+} > Cd^{2+} > Zn^{2+}$ and $Cd^{2+} > Cr^{3+} > Zn^{2+}$ on kiwi and tangerine. Rambabu *et al.* (2020) investigated the biosorption ability of date palm fruit bunch to remove hexavalent chromium (Cr^{6}) ions. Maximum biosorption of chromium removal efficiency was observed at 58.02% at 2 pH with 120 minutes of agitation time. Abatal *et al.* (2021) reported the removal of Pb, Cd, Co, and Ni ions by seeds powder of *Moringa oleifera.* The maximum sorption capacity was recorded of Pb followed by Co > Cd > Ni. *Nigella sativa* seed biomass used for sorption of lead (Pb) ions and the removal of lead from activated sludge waste and its decanted effluent confirm the efficiency of the biomass, which attained 97 % and 64 %, respectively (Addala *et al.*, 2018). Priyantha and Kotabewatta (2019) studied on the peel of edible fruit, *Atrocarpus nobilis* sorption capacity of Ni (II). The maximum adsorption of Ni (II) was observed on Freundlich model 12,048 mg kg^{-1}. Mallampati *et al.* (2021) used three natural materials, fruit peel (avocado, hamimelon, and dragon), as efficient renewable adsorbent to remove heavy metals. Dragon fruit peel showed the highest extraction efficiency toward alcian blue (71.85 mg/g) and methylene blue (62.58 mg/g). Hamimelon peel and avocado peel showed moderately extraction capacity Pb^{2+} (7.89 mg/g, 9.2 mg/g) and Ni^{2+} (9.45 mg/g, 4.93 mg/g) cation. Mohamed *et al.* (2019) studied on Ajwaa seed powder for the removal of selected heavy metals and observed that maximum removal efficiency in order 93.34%, 71.03%, 92.06%, 99.96%, 95.91%, and 36.13 % for ions Cd^{2+}, Cr^{3+}, Co^{2+}, Cu^{2+}, Pb^{2+} and Mn^{2+} respectively. Oboh and Aluyor (2008) investigated sour sop seeds to remove heavy metals, and the maximum biosorption capacity was achieved at 77.6%, 68.5%, 56.4%, and for ions 40.6% for ions Cu^{2+}, Ni^{2+}, Zn^{2+} and Pb^{2+}, respectively. The seeds of the bottlebrush were studied as an adsorbent removing of Cd (II), and maximum adsorption Capac was observed atrved 98% at pH 5 (Rao and Kashifuddin, 2014). Coelho *et al.* (2014) studied on cashew nut shell for the removal of Lead (II), Cromium (III) and Cadmium (II). The best condition for adsorption of ions Cd (II), Pb (II), and Cr (III) was observed at pH 5, adsorbent doses of 600 mg for 60 minutes of equilibrium time.

Stem used as an adsorbent

Lee *et al.* (2019) studied tea stem as a sorbent for removing methylene blue and the maximum adsorption capacity of tea stem powder was observed at 103.09 gm/g at pH 3.0-5.0. Zengin *et al.* (2018) investigated on vine stem and Turkish pine sawdust as a biosorbent for removing lather dye. The maximum adsorption capacity was 25.91 and 26.67 for vine stem and Turkish pine sawdust, respectively. Dried biomass of *Haloxylon recurvum* plant stems was used to remove methylene blue dye from an aqueous solution. The dye uptake by plant adsorbent increased with increased pH, contact time, and dye concentration. The maximum adsorption capacity was observed at 22.93 mg/gat the 8 pH (Hassan *et al.*, 2013). Schwantes *et al.* (2018) studied on the grape stem as an adsorbent for cadmium removal. The modified adsorbent exhibited Cd^{2+} reduction of 66% for E. NaOH, 33% for E. H_2O_2 and 8.3% for E. H_2SO_4.

Tree-based adsorbent used for removal of Cd (II) ions

Adsorbent of tree parts viz. leaves, stem, bark, seeds and stem were prepared for removal of Cd (II) ions. Adsorbent of Callistemon species tree seeds received maximum Cd (II) removal capacity 98% (Rao and Kashifuddin, 2014) and minimum received were 35.48% by *Cassia siamea* (John *et al.,* 2011) bark (Figure 3).

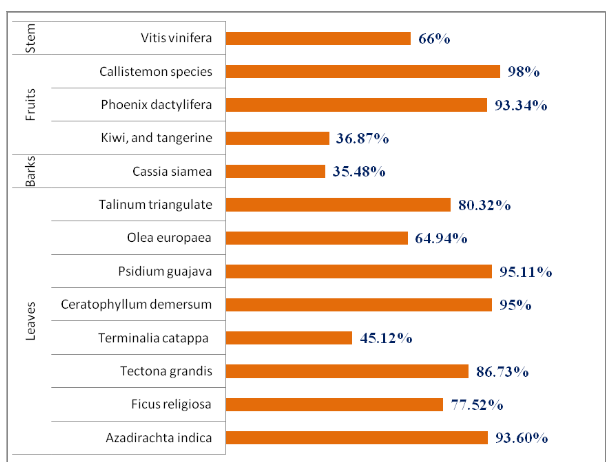

Fig. 3. The removal of Cd (II) ions by different tree species with adsorbent prepared by leaves, bark, seeds, and stem.

Tree-based adsorbent used for removal of Pb (II) ions

Adsorbent were prepared by leaves, seeds and bark of tree species for removal of Pb (II) ions. The seeds of *Nigella sativa* were maximum removal capacity 97% and minimum 40.30 % of *Pongamia pinnata* (Mamatha *et al.*, 2012) bark (Figure 4).

Tree-based adsorbent used for removal of Ni (II) ions

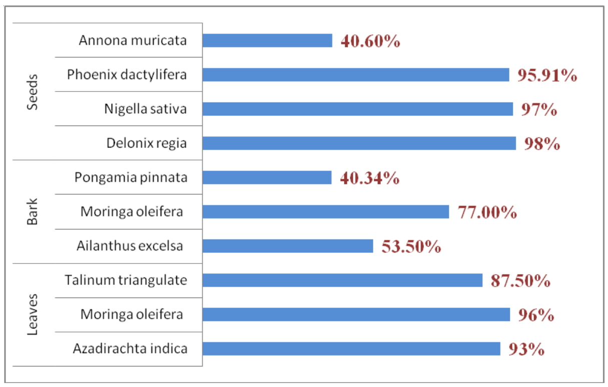

Fig. 4. Shows the removal of Pb (II) ions by different tree species with adsorbents prepared by leaves, bark, and seeds.

Tree species were used as adsorbent for removal of Ni (II). The maximum removal capacity of Ni (II) were recorded 98.02% by adsorbent of *Moringa oleifera* leaves (Reddy *et al.*, 2011) and minimum was 68.50% (Oboh and Aluyor, 2008) by adsorbent of Sour soap tree bark (Figure 5).

Equilibrium isotherm

An isotherm model explains the relationship between the amount of metal absorbed by the adsor-

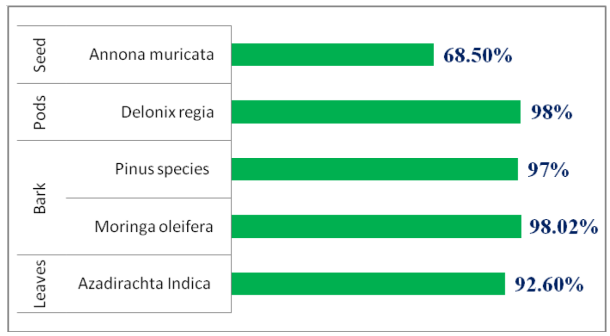

Fig. 5. Shows the removal of Ni (II) ions by different tree species with adsorbents prepared by leaves, bark, and seeds.

Table 1. Various equilibrium isotherm models was used in the adsorption process.

Metals	Adsorbents	Equilibrium model	Reference
Cd^{2+}, Pb^{2+}, Cr^{3+}	Cashew nut	Langmuir, Freundlich, Dubinin-Radushkevich (D-R)	Coelho *et al.* (2014)
Cd^{2+}	*Luffa cylindrical*	Langmuir, Temkin, Freundlich, Dubinin-Radushkevich (D-R)	Gongden *et al.* (2016)
Methylene blue, Ni^{2+}, Pb^{2+}	Avocado, Hamimelon, Dragon	Langmuir, Freundlich	Mallampati *et al.* (2015)
Ni^{2+}	*Artocarpus nobilis*	Langmuir, Freundlich	Priyantha and Kotabewatta (2019)
Pb^{2+}	*Nigella sativa*	Langmuir	Addala *et al.* (2018)
Cd^{2+}, Pb^{2+}, Ni^{2+}, Co^{2+}	*Moringa oleifera*	Langmuir, Freundlich	Abatal *et al.*(2021)
Pb^{2+}, Ni^{2+}	*Delonix ragia*	Langmuir, Freundlich	Festus *et al.*(2013)
Cr^{6+}	Date palm	Temkin	Rambabu *et al.*(2020)
Cd^{2+}, Cr^{3+} Ni^{2+}	Banana, kiwi, and Tangerine	Langmuir	Al-Qahtani (2015)
Hg^{2+}	Banana	Langmuir	Salamun *et al.*(2015)
Methylene blue dye	*Haloxylon recurvum*	Langmuir, Freundlich	Hassan *et al.*(2013)
Basic Blue 3 and Congo red dye	*Ananas comosus*	Langmuir, Freundlich, Temkin	Chan *et al.* (2016)
Methylene	Tea	Langmuir, Temkin	Lee *et al.* (2019)
Leather dye	Turkish red pine and Vine	Langmuir	Zengin *et al.* (2018)
Cr^{6+}	*Acacia nilotica*	Freundlich	Rani *et al.* (2005)
Pb^{2+}	*Ailanthus excelsa*	Langmuir, Freundlich	Waghmare and Chaudhari (2013)

bent and the metal concentration still present in the solution. Previously researchers used isotherm models to know the adsorption capacity of different metal ions, as shown in Table 1.

Conclusion

The adsorption process contributes significantly to the remediation process due to its ease in operation, selectively for metal ions, broad applicability in pollutant removal, and affordability. This review describes the tree-based adsorbent (leaves, stems, seeds, fruits, barks, etc.) used to remove heavy metals. Trees are available, planted, or naturally everywhere and easily provide natural adsorbent in the more considerable amount. It plays a significant role in improving water quality and environmental conditions.

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