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A review of fruit waste-derived sorbents for dyes and metals removal from contaminated water and wastewater

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ABSTRACT

This review article presents the application of various fruit wastes as effective and efficient sorbents to remove dyes and heavy metals from contaminated water and wastewater. Dyes and metals are described accordingly along with the existing technology available for their removal including membrane filtration, coagulation/flocculation, biological techniques, advanced oxidation processes, and adsorption. The characteristics and toxicity aspects of those pollutants are also provided. Then a specific insight related to the proven potentiality of sorbents especially derived from various fruit wastes such as banana peel, mango seed, and durian shell is explained deeper. These biosorbents are rich in particular organic compounds and functional groups in nature playing an important role in pollutants adsorption. Several modification processes such as carbonization, pyrolysis, and chemical activation using different chemical solutions like HCl, NaOH, and H_3PO_4 can be conducted to those fruit waste-based sorbents to activate their surface area for higher pollutant removal efficiency. To understand the adsorption mechanism, several prominent isotherm kinetics, adsorption modeling, and thermodynamic analysis are also discussed well here. Finally, based on the literature review on these raw and modified fruit waste-based sorbents, conclusions, challenges and future outlook have been critically drawn for obtaining better perspectives.

Keywords: Dyes; Heavy metals; Biosorption; Chemical impregnation; Carbonization

1. Introduction

The increasing number of industrial manufactures has resulted in increasing polluted water and wastewater streams containing dyes and heavy metals [1,2]. These pollutants have been generated in a huge amount due to their massive usage mainly in various manufacturing processes [2]. Various industries such as petroleum refinery, textiles, pulp and paper, pharmaceuticals, cosmetics, and paints release wastewaters containing synthetic dyes or heavy metals posing negative consequences to human health and other living creatures due to their carcinogenic and non-biodegradable characteristics [3–10]. Therefore, an effective and efficient way to increase the quality of contaminated water and wastewater before entering final disposal places is a must.

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To date, a number of treatment methods are available and have been implemented to remove dyes and heavy metals in wastewater solutions. Several techniques like coagulation/flocculation, flotation, biological ways, adsorption process, and membrane filtration can be considerable options [11–13]. Each of the methods aforementioned has its own advantageous and disadvantageous aspects. However, among them, the adsorption method is still considered a reliable mode due to low energy consumption, modest operation procedure, and high removal efficiency [14].

Then the increasing awareness of applying low-cost materials for creating effective adsorbents and keeping the environment sustainable has resulted in more attention to natural-based sorbent materials. The promising potentiality of sorbents derived from varying natural substances like zeolites, clay, chitosan, and red mud as well as agricultural and animal waste biomass has been reported widely [3,4,15]. Despite several articles already reviewed sorbents derived from low-cost natural materials, literature shows that the particular discussion about the use and valorization of different fruit waste-based sorbents to remove dyes and metals in water and wastewater is still limited. Therefore, this review article then specifically corroborates the relatively recent usage of fruit wastes such as apple peel, banana peel, durian shell, mango peel and many others for dyes and heavy metals removal. Authors also tried to analyze the scattered information available related to dyes and metals characteristics, their usage in assorted industries, existing technology available for removing these pollutants, adsorption isotherm, kinetics, modeling analysis, and the performance of different modified fruits waste-based sorbents along with some relevant challenges and future perspective of this area.

2. Dyes and metals in wastewater solutions

2.1. Dyes

The majority of dyes are complex organic molecules [8]. They are utilized in many edible and non-edible products due to their ability to improve the appearance of the products. It is estimated that more than a hundred thousand commercial dyes have been applied in various manufactures resulting in hundreds of tonnes per year of dyes disposal [16,17]. Therefore, their concentration in water and wastewater needs to be significantly reduced in the first place.

2.1.1. Dyes characteristics and application

In accordance with its solubility, color agents can be divided into two parts namely dyes and pigments. Dyes are natural or synthetic organic compounds that are hydro or oil-soluble while pigments are insoluble which remain in particulate forms [10].

Based on the source, dyes can be classified into two types, natural and synthetic dyes [10]. Natural dyes are made from mostly natural biomass such as plants and minerals. Plants-derived dyes are able to be generated from various plant parts including fruit, bark, leaf, and wood. Some natural dyes can also be produced from animals' body parts. All of those natural dyes have a high biodegradability level, so they are environmentally friendly. In contrast, synthetic dyes derived from organic or inorganic compounds have chemical structures of particular chromophore groups containing remain chemical substances such as mercury, lead, chromium, copper, and benzene [10,18]. Therefore, long exposure to untreated large dosages of synthetic dyes contained in wastewaters will cause harmful effects on both the human body and the ecosystem.

In terms of its chemical structure, the dye can be categorized into several groups as follows [8,19];

- Azo dye

Azo dye is synthesized from an aromatic amine with a chromophoric azo (–N=N–) group. This group is the largest percentage of synthetic dyes linked to the –OH or $-NH_2$ type auxochrome groups known as monoazo, diazo, or triazo dyes.

- Anthraquinone dye

This dye has general formula derived from anthracene and chromophore group called quinone nucleus which has a special ability to bind both hydroxyl and amino groups.

- Indigo dyes

It contains selenium, sulfur, and oxygen homologous compounds of indigo blue resulting in hypochromic orange or turquoise color.

- Xanthene dye

This dye has an intense fluorescence making it attractive.

- Phthalocyanine dye

Phthalocyanine dye can be produced using the reaction of dicyanobenzene in the presence of certain metals such as Cu, Ni, Co, Pt.

- Nitrated and nitrosated dyes

They have a specific molecular structure characterized by the presence of a nitro group ($-NO_2$).

2.1.2. Dyes and industry

The relationship between dyes and industry is undeniably strong. In certain food industries, natural dyes, such as pumpkins and green leaves are still used to some extent. However, the use of synthetic dyes mostly derived from petroleum is significantly increasing due to their bright and stable colors [10]. In the pharmaceutical industry, dyes are also applied to give both a better appearance and identification of medicinal products. Colored medicine products will be easier to be recognized and, at the same time, increase the vigilance of fake medicine widespread [20].

Furthermore, in the cosmetics industry, synthetic dyes are used in high dosage and more varieties due to economical and long-lasting brightness aspects towards manufacturing operating conditions like light, heat, and pH [21]. Cosmetic products including lipstick, eyeliner, and hair dyes along with other basic needs such as shampoo and soaps need dyes to beautify their outward. Another industry using massive amounts of synthetic dyes is the textile industry. As part of the fast-growing industries, textile manufacturers need dyes for dry and wet fabric textile products to make them eye-catching [19]. It is estimated that the global production of textile dyes is more than 10,000 ton/y, and about 100 ton/y of dyes disposals are released into open water [22,23]. Unfortunately, most of those synthetic dyes are highly toxic and low biodegradable causing challenging efforts for their total or partial elimination.

Among a wide variety of dyes, some dyes have been more commonly applied in manufacturing processes such as Methylene blue ($C_{16}H_{18}ClN_3S{\cdot}3H_2O$) and Malachite green ($C_{23}H_{25}N_2$). Both of them are cationic dyes that has been used massively in manifold industrial sectors despite their hazardous nature affecting the wellbeing of living creatures including human lives [24]. Other prevalent dyes employed in manufacturing processes are then tabulated in Table 1.

2.2. Heavy metals

Heavy metals are non-biodegradable and toxic elements tending to accumulate in living organisms. Water and wastewater contaminated by heavy metals have to have sufficient treatment before they are allowed to enter the open environment. Several metals are commonly used in many industries and their toxicity effect is described in Table 2.

2.2.1. Mercury

As a neurotoxic heavy metal, mercury (Hg) exposure threatens human lives. This corrosive substance could also give a damaging effect on industrial machine installation such as aluminum heat exchangers as it can accumulate in the form of liquid or solid particles on it [25]. Mercury-related manufacturing sectors like paper, pulp, ore, and batteries production are disposing of significant levels of mercury in the wastewater streams. Long exposure to mercury will lead to serious health damages like neurological disorders, paralysis, and blindness. Hence, its concentration in wastewater must be reduced until reaching the permissible limit set by the relevant institution. European Union (EU) set a safe standardized value of mercury by 0.001 and 0.005 mg/L in drinking water and wastewater, respectively [26,27]. This concentration level could only be achieved through proper treatments, so treated water and wastewater could be reused or exonerated to the final disposal places.

Table 1
Several commonly used dyes in assorted industries

Name of dye	Molecular formula	Source	Colour	Application
Carotenoids	$C_{10}H_{56}$	Natural; potatoes, pumpkins, tomatoes	Red, yellow, orange	Foods; drinks, medicine
Chlorophyll	$C_{55}H_{72}MgN_4O_5$	Natural; green plants	Green	Foods; drinks, medicine
Anthocyanin	$C_{15}H_{11}O^+$	Natural; berries fruits, tomatoes	Red, purple, blue	Foods; drinks, medicine
Turmeric/curcumin	$C_{21}H_{20}O_6$	Natural; stem underground	Yellow, orange	Foods; drinks, medicine, cotton fabric
Bugs	–	Natural; bugs	Red	Foods; yogurt, juice
Tartrazine	$C_{16}H_9N_4Na_3O_9S_2$	Synthetic; coal tar	Yellow, green	Foods; candy, soft drinks, gum, honey
Brilliant blue FCF	$C_{37}H_{34}N_2Na_2O_9S_3$	Synthetic; coal tar	Dark blue, light blue	Foods; yogurt, cheese, butter
Amaranth	$C_{20}H_{11}N_2Na_3O_{10}S_3$	Synthetic; coal tar, petroleum by products	Dark red	Foods; cosmetics, fibre, paper, leather
Allura Red AC	$C_{18}H_{14}N_2Na_2O_8S_2$	Synthetic; petroleum by products	Red, orange	Foods; candy, chocolates, soft drink, cosmetics, medicine
Brilliant black	$C_{28}H_{17}N_5Na_4O_{14}S_4$	Synthetic; petroleum by products	Black	Foods; mustards, ice cream, jams, medicine
D&C Black No. 2	C	Synthetic; petroleum oil	Black	Cosmetics; mascara, lipstick, soap, shampoo, hair dyeing
FD&C Yellow No. 5	$C_{16}H_9N_4Na_3O_9S_2$	Synthetic; trisodium salts	Yellow	Cosmetics; medicine, foods
Patent Blue No. 5	$C_{27}H_{33}N_2O_7S_2^+$	Synthetic; triphenylmethane backbones	Blue	Foods; medicine
Black Reactive 5	$C_{26}H_{21}N_5Na_4O_{19}S_6$	Synthetic; coal tar, petroleum by products	Black	Textile; paper
Phthalocyanine	$C_{32}H_{18}N_8$	Iron phthalocyanine	Blue, green	Textile; paint

Table 2
Assorted heavy metals contained in industrial wastewaters

Name of metal	Permissible limit	Industries water waters containing metal	Toxicity effect
Mercury	0.001 mg/L	Pharmaceutical, paper, pulp, ore, and batteries production	Neurological disorder, paralysis, blindness, weak immune system
Lead	0.05 mg/L	Battery, ammunition, bronze, pipe, ceramic, roofing materials, and glass production	Brain damage, anemia, anorexia, vomiting, and disease of circulatory, nervous systems and mental illness
Arsenic	0.01 mg/L	Glass, textile, paper, insecticides, phosphate fertilizers, mining, and coal combustion	Lung, bladder, and kidney cancer, neurological disorder, liver tumor muscular weakness, and nausea
Copper	1.3 mg/L	Medical equipment, fertilizer, paints, pigments, tanneries, and cleaning products	Liver damage and lung cancer, insomnia, osteoporosis, heart disease, cancer, migraine headaches, and seizures
Cadmium	0.003 mg/L	Fertilizer, batteries, metal plating, power plants, solid waste, mining, smelting, and fuel combustion	lung cancer and kidney failure, renal disturbances, lung fibrosis, and bone lesions
Chromium	5 mg/L	Steel fabrication, dyes, textile industries, and ceramics	cancer, lung tumor, hemorrhage, mutagenic, teratogenic, epigastric pain nausea, vomiting, and severe diarrhea
Nickel	0.015 mg/L	Batteries, coinage, electroplating, mining, refining, glass, and paints	Dermatitis, chronic asthma, and cancer, chronic bronchitis, reduced lung function, and lung cancer
Zinc	5.0 mg/L	Mining, galvanization, stabilizers, steel production, and coal incineration	Gastrointestinal distress, nausea, lethargy, neurological signs, and loss of appetite

2.2.2. Lead

Lead (Pb) generally exists in the wastewater streams of battery manufacturing, ammunition, bronze products and pipe, ceramic and glass industries. This metal has two main oxidation states commonly named Pb(IV) and Pb(II) and can cause detrimental effects on human health related to brain damage, damaged circulatory and nervous systems [28]. Lead accumulated in soils could remain for hundreds of years negatively affecting plant's photosynthesis process and ecosystem food chain [29]. Due to its severe detriments, World Health Organization (WHO) then set the permissible level of lead by 0.05 mg/L [30].

2.2.3. Arsenic

Arsenic (As) is a metalloid element that is highly toxic especially in its inorganic form, and it can exist in different oxidation shapes [31,32]. These metal ions are mostly used as alloying agents and can be generated by assorted industries especially coal combustion. Long exposure through ingestion of arsenic-contaminated water results in the dysfunctional lung, bladder organs, and kidney cancer [33]. Therefore, WHO stated that the permissible limit of As in drinking water is just 0.01 mg/L [34].

2.2.4. Copper

Copper is accumulated in the form of ore deposits formed by hydrothermal processes, and it can be found in nature mostly in volcanic and sedimentary rock [35]. It is applied in many industries including paints, pigments, electrical wire, and fertilizers [36]. Even though some research claimed that this type of metal is useful for various medical and agricultural purposes to some degree, prolonged exposure to copper in relatively high concentrations will increase the chance of having life-threatening illnesses such as liver damage and lung cancer. Then the maximum permissible limit of copper was ruled by the United States Environmental Protection Agency (USEPA) at 1.3 mg/L [37].

2.2.5. Cadmium

Cadmium (Cd) is a less essential metal element, but it has a high solubility in water resulting in a higher chance to accumulate in the watery environment [38]. The contamination of cadmium ions can occur in the wastewater streams of certain industries such as fertilizer production, metal plating, power plants, mining, smelting, and fuel combustion. Long-term exposure beyond safe limitation to this metal would cause cancer. It is then important to set its allowable cadmium content, and WHO set the permissible value of this metal by 0.003 mg/L [39].

2.2.6. Chromium

Chromium (Cr) metal ions can be detected in the wastewater streams of several manufacturing plants such as steel fabrication, textile industries, and ceramics products [40]. More than one-third of carcinogenic chromium substance contained in industrial effluent is in the form of Cr(VI) or Cr(III) chromium in which Cr(VI) is found to be much more hazardous than Cr(III) [41]. For drinking water, EPA stated that the permissible limit of Cr(VI) is 0.1 mg/L while WHO set the limit of Cr(III) by less than 5 mg/L [14].

2.2.7. Nickel

Nickel (Ni), a silver-white color, hard and squashy metal type, is usually used in manufacturers regarding batteries, coinage, electroplating, mining, refining, glass, and paint production [42–44]. An adequate wastewater treatment technique is essential as overwhelming exposure to nickel could cause malignant impacts on human health like chronic asthma and cancer. For this reason, USEPA has stated the permissible standard of nickel in drinking water by 0.015 mg/L [43].

2.2.8. Zinc

Zinc (Zn) is a mineral and part of heavy metals commonly dispersed in water and wastewater streams. Zinc is generally used in mining, galvanization, and coal incineration. The excess amount of this element could stimulate harmful side effect like lethargy, neurological signs, and loss of appetite, WHO then standardized the maximum allowable limit of zinc by 5.0 mg/L [45,46].

3. Existing treatment methods for dyes and heavy metals removal

3.1. Membrane filtration

In the last decades, membrane technology has been applied widely to remove various pollutants to produce treated water and wastewater with high purity levels. In terms of particle size passing membrane pore, the membrane can be categorized into several types such as microfiltration, ultrafiltration, and nanofiltration. Compared to other treatment methods, membranes filtration has shown great work for their high efficiency including for dyes and metals depletion [47–50]. Despite its well-known practical operation, highly flexible design, and less sludge production, the membrane system needs higher maintenance and operational cost to keep its performance well [51]. The main problem of membrane technology is the fouling phenomenon which can occur during the filtration process. Hermia's models comprising complete blocking, standard blocking, intermediate blocking, and cake filtration models than could be used to analyse fouling mechanism [52] as illustrated in Fig. 1.

3.2. Advanced oxidation processes

Advanced oxidation processes (AOPs) are part of methods to eliminate pollutants from wastewater due to their

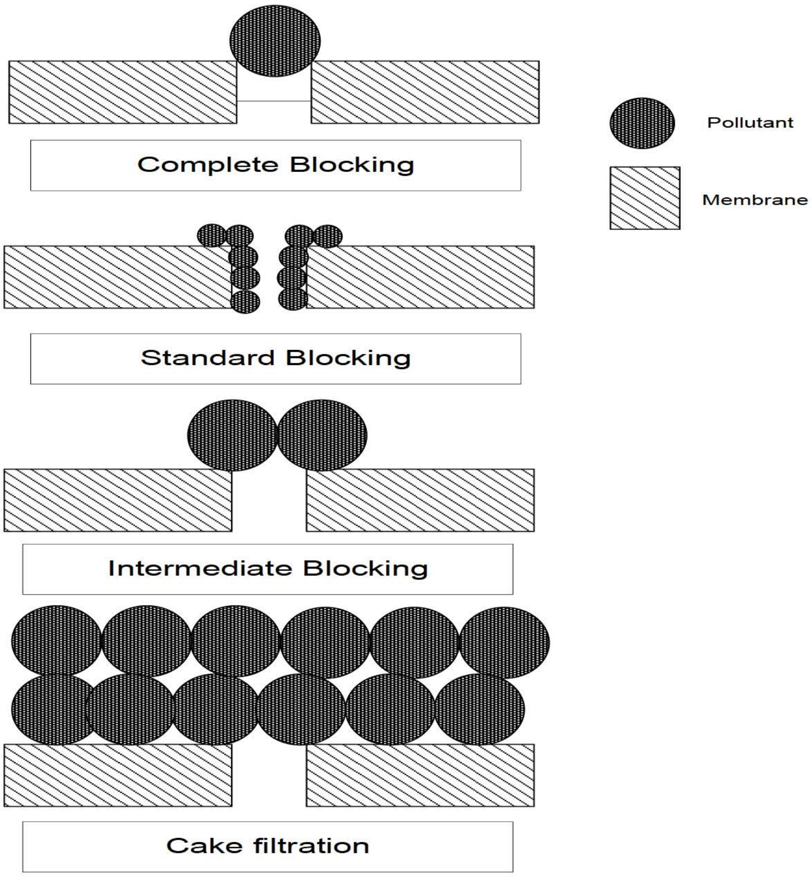

Fig. 1. The mechanism of membrane fouling.

excellent capability in organic compounds mineralization due to hydroxyl radical production [53]. Several modes of performing AOPs are Fenton (H_2O_2/Fe^{2+}), Fenton-like (H_2O_2/Fe^{3+}), electron beam irradiation, sonolysis, electrochemical oxidation, photo-assisted Fenton ($H_2O_2/Fe^{2+}/Fe^{3+}/UV$), and photocatalytic mechanisms [54–57]. Some studies reported the efficacy of this method regarding dyes elimination in contaminated water [58,59]. However, there are also several shortfalls of AOPs that have to be considered such as the need for costly chemicals, post-usage impacts, and the production of useless sludge and other by-products [60].

3.3. Biological process

In terms of biological methods, the involvement of microorganisms is a must as the existence of these living creatures enables the degradation process of diluted pollutants contained in wastewater. Despite some possible drawbacks such as more time and place needed for the biological reaction to occur, this process offers more environmentally friendly characteristics, great removal efficiency, and lower operational cost [61]. Biological agents which can be aerobic (with oxygen), anaerobic (without oxygen), or combined aerobic–anaerobic microorganisms are abundantly available in nature including fungi and bacteria which are reliable to eradicate metal ions or dye molecules in contaminated solutions [62–65].

3.4. Coagulation and flocculation

Coagulation and flocculation refer to the application of flocculants and coagulants such as aluminum and ferric chloride [66]. This process involves several influential parameters playing important roles such as initial pollutant concentration, coagulants and flocculants dosage, and pH. In this method, there are some fundamental points that have to be concerned such as how to destabilize, entrap, and aggregate colloids related to the pollutants flocks by filtration stage (Fig. 2) [67]. Despite the efficacy, there are also some disadvantages of using this method including costly reagents and massive generation of toxic sludge along with its discharge concern that need further sustainable efforts [68].

3.5. Hybrid method

Since each treatment technique has its own advantageous and disadvantageous points, some literature proposed hybrid methods which are considered as a better breakthrough to reach higher removal efficiency and friendlier environmental aspects. Several studies have reported their work on a hybrid of two or more treatment methods with outstanding results. Treating raw industrial wastewaters which especially have high loading of harmful pollutants using a sole membrane filtration system, for example, can lead to a higher membrane fouling rate. It then can be risky without combining membrane system with pre-treatment process as high fouling rate can cause shorter life-span and costly maintenance funding [69–71]. Therefore, some studies integrated their wastewater treatment methods. The hybridization of electrocoagulation, ceramic microfiltration membrane, and ozone techniques was also investigated for treating oily wastewater with high recalcitrant loading [72]. It reported a drastic increase in chemical oxygen demand removal efficiency from 18% to more than 53%. Combining ultrafiltration membrane with photocatalytic or adsorption method was found effective to increase pollutant removal efficiency and decrease fouling rate [71,73]. In addition, the integration of other techniques including photo-oxidation, adsorption, condensation, and electrochemical to remove various dyes and heavy metals and dyes also showed more satisfactory outcomes than those of using sole treatment work [74,75].

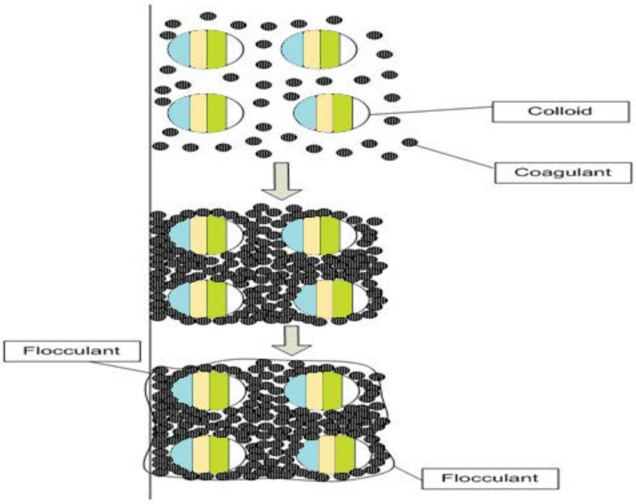

Fig. 2. The mechanism of coagulation/flocculation.

3.6. Adsorption and biosorption

Theoretically, adsorption is atoms or ions accumulation process to sorbent pores and surface creating a particular layer consisting of dyes, metal ions, soil particles or other organic and inorganic pollutants (Fig. 3) [76,77]. Adsorption is a well-known water treatment technique notable for its simple and reliable output with sufficient removal efficiency, less energy consumption and less amount of chemicals. However, it has also some drawbacks such as costly material preparation and regeneration process [78]. Due to the need of keeping environmental sustainability well, there is increasing popularity of biosorbent which has effective, eco-friendly, and economical perspectives [4]. The adsorption of contaminants from wastewater using different sorbent types such as commercial sorbents made from graphene and activated carbon; natural sorbents made from clays and zeolites; and organic sorbents made from plant or animal biomass has indicated remarkable competitive outcomes [4,79–82].

Furthermore, the term biosorption could be defined as the ability of particular biomass to bind and remove targeted pollutant ions or molecules from an aqueous solution [83]. The abundance, availability and low-cost aspects have triggered people to explore and valorize unwanted

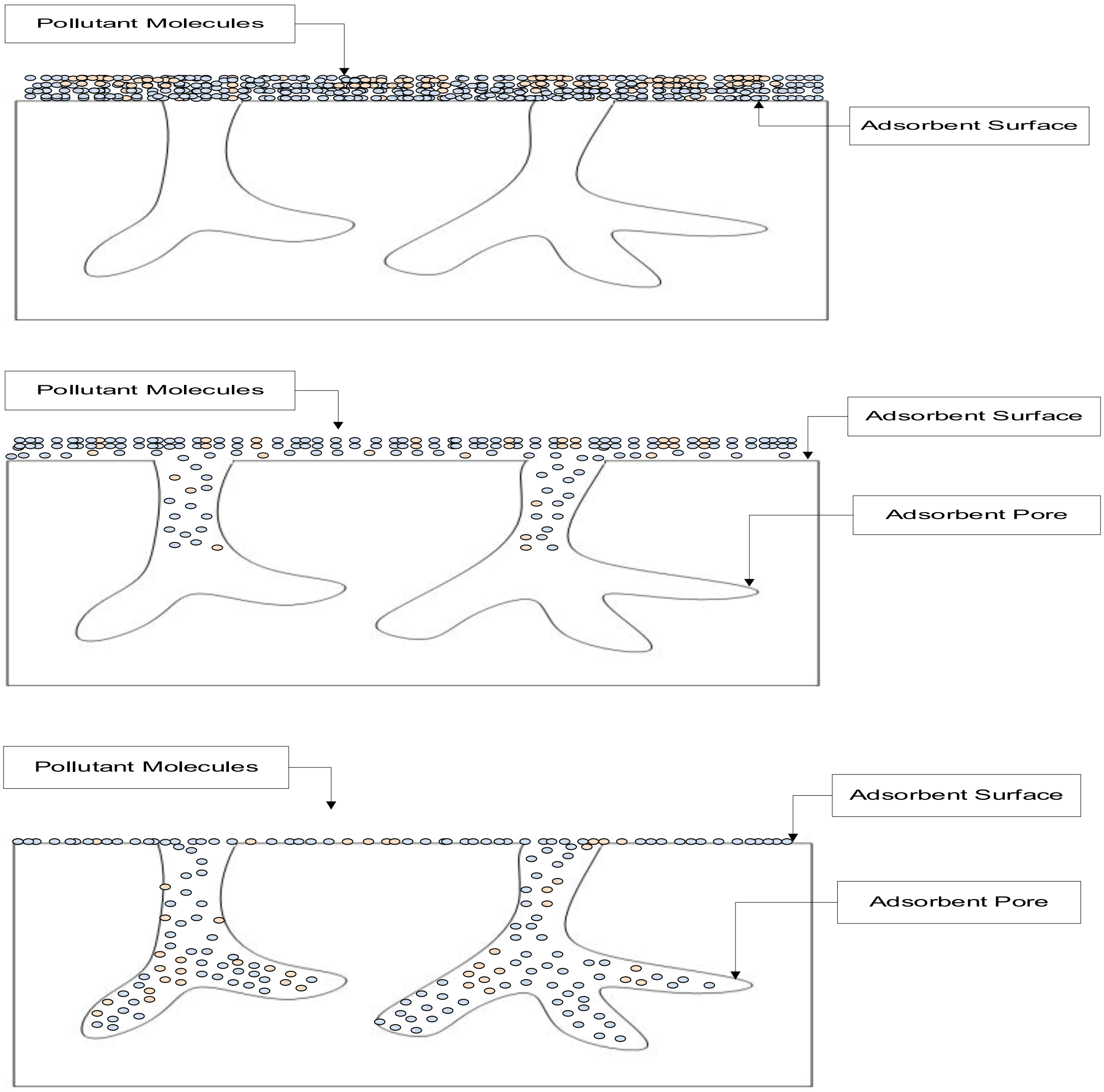

Fig. 3. The mechanism of adsorption.

biomass to become effective biosorbent. Hence, the potentiality of various plant wastes including fruit remains for reducing both dyes and heavy metals ions were further assessed. Generally, biosorption consists of chemisorption, physisorption, microprecipitation and oxidation/reduction [84]. Chemisorption relates to ion exchange, chelation and complexation or coordination processes while physisorption correlates to electrostatic interaction and van der Waals forces. Surface functional groups on the biosorbent particle have an important function for the binding pollutants in the chemisorption stage while ion exchange involves electrostatic interactions between negatively charged biosorbent cell wall and the cations contained in the solution [85]. The chelation process refers to the attachment of a ligand with a metal ion resulting in a ring structure formation creating a barrier for the attached mineral to enter the unwanted chemical reactions [1]. For metal removal, it connects to the complexation process by forming a complex with the active groups of the cell wall on the cell surface of the biosorbent. The bonding formed could be covalent or electrostatic, and the compounds may be neutral, negatively or positively charged. Different from the chemisorption aforementioned, physisorption is a non-specific reversible process involving weaker van der Wall's forces [86]. Pollutant molecules form a monolayer over the surface of the biosorbent with lower activation energy.

There are several factors affecting the removal efficiency of pollutants in the solution such as particular pollutant properties, sorbent properties like porosity and surface area, and operating conditions such as pH, temperature, sorbent dosage, and initial concentration [8,87]. The tendency of particular variables is critical to be considered to create a supportive environment for a better biosorption process. Acidic pH range from 3 to 5, for example, tends to be more favorable to maximize metals or dyes removal efficiency than other pH values. This correlates to a better chance of electrostatic attraction between positively charged biosorbent surface and negatively charged pollutant particles [88]. However, this may vary to specific types of pollutants and sorbents which have had surface structure change due to the modification process. Initial concentration also holds a considerable role where the higher amount of initial pollutant will lead to its lower removal efficiency [89]. Increasing biosorbent dosage could mainly increase removal efficiency to some extent while the temperature may have a different effect on the removal efficiency depending on the characteristics of targeted pollutants [88,90]. This may be caused by the natural resistance of certain pollutant particles and sorbent materials to temperature levels. In order to optimize the efficacy of biomass, several modifications can be conducted. The options are mainly carbonization, chemical activation, pyrolysis or hybrid activation modes as modified biosorbent will have more active surface related to a better adsorption capacity [91–93].

4. Adsorption isotherm models

In an adsorption process, there are several parts interacting with each other named sorbent (a solid phase) and a liquid phase (generally water or aqueous solution) containing pollutants that are attracted into sorbent surface due to the affinity of the sorbent. This process will happen continuously until an equilibrium condition is established between the amount of solid-bound pollutants and its portion remaining in the solution at an equilibrium concentration [3,83]. It is fundamental to study the quantification of pollutant and sorbent interactions to examine targeted pollutant uptake by the sorbent. This can be expressed as batch equilibrium isotherm related to mechanistic models or empirical equations. While mechanistic models are based on the mechanism of adsorption that could predict the adsorption behavior during experimental work, empirical models for a single solute system refers to the way the distribution of pollutant particles between the liquid phase and sorbent surface at equilibrium during adsorption. There are several reliable isotherm models that could be applied to understand the adsorption mechanism.

4.1. Freundlich isotherm model

This model assumes that the adsorption process occurs on the heterogeneous surface having unequal available sites with different adsorption energies as formulated in Eq. (1) [94,95].

$$\ln q_e = \ln k_f + \frac{1}{n}\left(\ln C_e\right) \tag{1}$$

where q_e, C_e, k_f, and n are the amount of pollutants adsorbed per unit of the sorbent at equilibrium time (mg/g), the equilibrium of pollutant concentration in the solution (mg/L), isotherm constant, and the adsorption intensity, respectively. Additionally, the adsorption isotherm is linear when $n = 1$, favorable when $n < 1$, and unfavorable when $n > 1$ [3].

4.2. Langmuir isotherm model

This model describes that the adsorption happens onto an ideal homogeneous uniform surface where all of the sites on the sorbent surface are equivalent. General linearized Langmuir isotherm can be written as follows [94].

$$\frac{C_e}{q_e} = \left(\frac{1}{K_L q_m}\right) + \frac{C_e}{q_m} \tag{2}$$

where q_m and K_L are maximum adsorption capacity (mg/g) and Langmuir constant, respectively.

4.3. Dubinin–Radushkevich isotherm model

This model implies the adsorption mechanism related to Gaussian energy distribution onto a heterogeneous sorbent surface. Then the linearized form of this model could be expressed as follows [96].

$$\ln q_e = \ln q_m - \beta\varepsilon^2 \tag{3}$$

where β and ε are a constant related to adsorption energy and the Polanyi potential related to the equilibrium concentration, respectively. The value of ε can be obtained from Eq. (4).

$$\varepsilon = RT\ln\left(1 + \frac{1}{C_e}\right) \tag{4}$$

where R is the gas constant (8.314 J/mol K) and T is the absolute temperature (K).

4.4. Temkin isotherm model

Temkin isotherm model considers the influence of indirect interaction among pollutants contained in the liquid media during the adsorption process along with the assumption that the adsorption heat of all molecules available in the layer would decrease linearly with the increase in the surface coverage [97]. The linearized formula for the Temkin isotherm model can be written as follows [98]:

$$q_e = \frac{R_t}{b}\ln K_T + \frac{R_t}{b}\ln C_e \tag{5}$$

where b and K_T are Temkin constant regarding the heat of sorption (J/mol) and Temkin isotherm constant (L/g), respectively.

4.5. Elovich isotherm model

Elovich isotherm model concerns the principle of kinetics assuming adsorption sites that would escalate exponentially with the adsorption rate implying a multilayer adsorption process [97]. The following equation can express the Elovich isotherm model well.

$$\ln\frac{q_e}{C_e} = \ln K_e q_m - \frac{q_e}{q_m} \tag{6}$$

5. Adsorption kinetic modeling

Adsorption kinetics and mechanism are pivotal to understand process design in adsorption. Kinetic studies will provide information on the predicted amount of adsorbed pollutants and other related aspects such as external mass transfer, intraparticle mass transfer, and solute diffusivity. Several kinetic models including pseudo-first-order, pseudo-second-order, intraparticle diffusion, liquid film diffusion, and double-exponential models are available to study adsorption kinetics and mechanism.

5.1. Pseudo-first-order

The pseudo-first-order reflects the occupation rate of adsorption sites in the proportional rate to the number of unoccupied sites. The linearized integral form of the pseudo-first-order kinetic is stated as follows [Eq. (7)] [94,95,99,100].

$$\log\left(q_e - q_t\right) = \log\left(q_e\right) - \frac{K_1}{2.303}t \tag{7}$$

where q_t, K_1, and t are pollutants adsorbed at a specific time (mg/g), equilibrium rate constant of pseudo-first-order adsorption (min^{-1}), and time (min), respectively.

5.2. Pseudo-second-order

The pseudo-second-order kinetic model can be linearized as following [101,102].

$$\frac{t}{q_t} = \frac{1}{K_2 q_e^2} + \frac{1}{q_e}t \tag{8}$$

where K_2 is the pseudo-second-order rate constant (g/mg min) that can be estimated by plotting t/q_t vs. t. Thus the constant K_2 can be used to obtain the initial sorption rate (h) at $t = 0$, as follows.

$$h = k_2 q_e^2 \tag{9}$$

The values of K_2, h, and q_e can be calculated by the plot of t/q vs. t.

5.3. Intraparticle diffusion

This model is used for adsorption mechanism identification specifically for design purposes [16]. For most adsorption processes, the uptake varies proportionally with $t^{1/2}$ rather than with contact time as represented by the following equation [94].

$$q_t = K_{\mathrm{id}} t^{0.5} \tag{10}$$

where K_{id} and $t^{0.5}$ are is the rate constant of intraparticle diffusion (mg/g min$^{0.5}$) and the square root of time (min), respectively. Plotting q_t vs. $t^{0.5}$ gives a linear relationship. Then K_{id} value can be decided from the slope of the plot.

5.4. Liquid film diffusion

In liquid and solid adsorption system, the rate of accumulated solute in the solid phase is equal to that of solute transfer and can be written as follows [94,103].

$$\ln\left(1 - \frac{q_t}{q_e}\right) = -K_{\mathrm{fd}}t \tag{11}$$

where $\ln(1 - q_t/q_e)$ and K_{fd} are the fractional attainment of equilibrium and the film diffusion rate constant, respectively. By plotting $\ln(1 - q_t/q_e)$ vs. t, it forms a linear relationship. Then K_{fd} can be obtained from the slope of the plot.

5.5. Double-exponential

A double-exponential function can be written as follows [104].

$$q_t - q_e = \exp\left(-K_1 t\right) - \exp\left(-K_2 t\right) \tag{12}$$

In the condition of $K_1 >> K_2$, the rapid process could be assumed to be negligible on the overall kinetics, and the linearized form of the equation can be written as follows:

$$\ln\left(q_e - q_t\right) = -K_2 t \tag{13}$$

where K_1 and K_2 are diffusion parameters of the rapid and the slow step (min^{-1}), respectively. Plotting $\ln(q_e - q_t)$ vs. t will give a linear relationship, where K_2 can be obtained from the slope of the plot.

6. Thermodynamics analysis

The thermodynamic analysis is fundamental to assess the effect of temperature on adsorption capacity and to understand the reaction changes during the adsorption process through specific parameters namely Gibbs free energy ($\Delta G°$), enthalpy ($\Delta H°$), and entropy ($\Delta S°$). The following equations are commonly used in thermodynamic studies [105].

$$\Delta G° = \Delta H° - T\Delta S° \tag{14}$$

$$\log\left(\frac{q_e}{C_e}\right) = \frac{\Delta S°}{2.303R} + \frac{-\Delta H°}{2.303RT} \tag{15}$$

where q_e and C_e are the solid-phase concentration at equilibrium (mg/L) and equilibrium concentration in solution (mg/L), respectively.

7. Modified fruit waste-based sorbents

As specified aforementioned, this review article is mainly inclined to gather the relatively recent exploration of various fruit wastes such as orange peel, apple peel, banana peel, durian shell, and many others for dyes and heavy metals removal. While their performance is affected by particular sorbent properties, operating condition and physicochemical behavior [3,4], initial modification techniques such as drying, carbonization, pyrolysis, calcination, chemicals impregnation or hybrid activation systems can also significantly boost their ability enabling higher adsorption capacity (Fig. 4) [90,91,106,107]. Further, the exploration of fruit waste-based sorbents for adsorbing dyes and heavy metals is tabulated in Table 3.

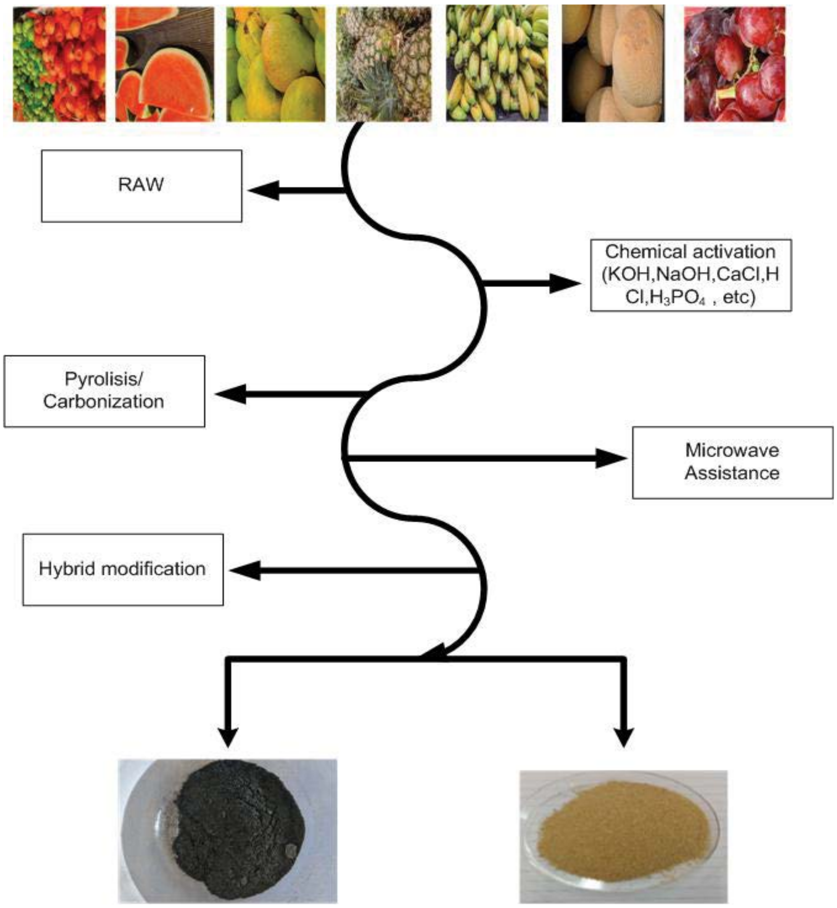

Fig. 4. Various modification techniques for valorizing fruit waste-based sorbents.

7.1. Orange peel

Orange fruit as part of citrus species is rich in healthy phytochemicals. The peel of this fruit is not considered as a consumable part leading to a massive disposal. The valorization process then is beneficial to do as the orange peel contains some substances useful for adsorption like pectin, lignin, and hydroxyl functional groups [108]. Bediako et al. [109] created AC synthesized from orange peel to adsorb different types of prevalent dyes from laboratory organic wastewater named cationic Methylene blue and anionic methyl orange. Orange peel was previously activated by chemical impregnation using several reagents such as H_2SO_4, NaOH, KOH, $ZnCl_2$, and H_3PO_4 followed by carbonization. Amongst several isotherm models they employed, the Langmuir model tended to have a better estimation of the maximum equilibrium uptakes close to the experimental data. They stated that adsorption capacities were about 1,012.10 ± 29.13 for $ZnCl_2$-AC (methyl orange), 339.82 ± 6.98 for $ZnCl_2$-AC (Methylene blue), and 382.15 ± 8.62 mg/g for KOH-AC (Methylene blue). Overall, the output showed an excellent removal efficiency of more than 99% for Methylene blue. Another study examined Fe-modified hydrochar made from orange peel for adsorbing Brilliant black dye [110]. By applying response surface methodology (RSM), they investigated the role of treatment time, dye concentration, sorbent dosage, and temperature, and determined the prominent mechanisms through kinetics analysis. The orange peel which was initially treated by hydrothermal carbonization at 200°C for 8 h was embedded with magnetite nanoparticles. Theoretically, it resulted in around 99% of dye removal efficiency with particular optimum conditions of dye initial concentration 6.08 mg/L, treatment time 26.30 min, temperature 44.79°C and sorbent dosage 2.27 g/L. Langmuir's model then showed a better congruence to the experimental data with a maximum adsorption capacity of 10.49 mg/g. Furthermore, Congo red dye removal from aqueous media using cationic modified orange peel sorbent was investigated by considering temperature [111]. Then it was found that the maximum adsorption capacity at different temperatures of 298, 308, and 318 K were 107, 144, and 163 mg/g, respectively. From this information, we could assume that higher temperature provides for better adsorption circumstances suitable for Congo red dye characteristics. The next study developed a quaternary amine-modified orange peel-based-sorbent to adsorb Reactive red 120 dye [112]. They reported pseudo-second-order kinetic and Langmuir isotherm as the best fit models to predict adsorption mechanism with regression coefficient close to 1. In accordance with the pH solution, it was revealed that increasing pH could decrease the adsorption capacity

Table 3
Fruit waste-based sorbents for dyes and heavy metals removal

Adsorbent material	Modification technique	Adsorbate/adsorbent capacity or removal efficiency	Isotherm model/ thermodynamic	Optimum operating conditions	References
Orange peel	ZnCl activation – carbonization	Methylene blue dye	Langmuir and Sips	99%	[109]
	Fe activation – carbonization	Brilliant black dye	Langmuir	10.49 mg/g	[110]
	Cationic modification	Congo red dye	Langmuir	163 mg/g	[111]
	Quarterly amine activation	Reactive red 120	Langmuir	344.8 mg/g	[112]
	NaOH activation	Crystal violet dye	Langmuir	27.17 mg/g; 97.5%	[113]
		Methylene blue dye		25.87 mg/g; 99.2%	
	Pyrolysis-chemical impregnation	Malachite green dye	–	28.5 mg/g	[114]
	Carbonization	Cu(II)	Temkin	94%	[115]
	–	Cu(II)	Freundlich	63 mg/g	[116]
	KOH activation – pyrolysis	Cd(II)	Langmuir	28.7 mg/g	[186]
		Cr(III)		30.11 mg/g	
		Co(II)		45.44 mg/g	
Banana peel	Chemical impregnation	Reactive black 5 dye	Langmuir	211.8 mg/g	[120]
	$FeSO_4$ activation – carbonization	Methylene blue dye	Langmuir	862 mg/g	[119]
	Acid activation	Rhodamine B	Langmuir	9.5220 mg/g	[121]
	H_3PO_4 activation – hydrothermal	Ni(II)	Langmuir	320 mg/g	[44]
	H_3PO_4 activation – pyrolysis			488 mg/g	
	Hydrothermal – calcination	Cu(II)	Langmuir	49.5 mg/g	[79]
		Pb(II)		45.6 mg/g	
		Cd(II)		30.7 mg/g	
		Cr(VI)		25.2 mg/g	
	Acid activation	Cr(IV)	Langmuir–Freundlich	88%	[122]
Mango peel	H_2SO_4 activation	Methylene blue dye	Langmuir	277.8	[128]
	Drying	Cr(II)	Freundlich	98.039 mg/g	[131]
		Cr(VI)		66.666 mg/g	
Mango seed	Drying	Malachite green dye	Langmuir–Freundlich	22.8 mg/g; 96%	[129]
	Chemical activation	Crystal violet dye	Langmuir	352.79 mg/g	[126]
Jackfruit leaf	Drying	Malachite green dye	Freundlich	80%	[133]
	KOH activation	Cr(VI)	Langmuir–Freundlich	62.7 mg/g	[136]
	H_3PO_4 activation			68.37 mg/g	

(Continued)

Table 3 Continued

Adsorbent material	Modification technique	Adsorbate/adsorbent capacity or removal efficiency	Isotherm model/ thermodynamic	Optimum operating conditions	References
Jackfruit peel	Carbonization	Cu	–	99.84%	[135]
	EDTA activation	Cr(III)	Langmuir	41.67 mg/g	[137]
		Ni(II)		52.08 mg/g	
	SDS activation	Cr(III)		26.25 mg/g	
		Ni(II)		20.88 mg/g	
	NaOH activation	Cr(III)		37.04 mg/g	
		Ni(II)		27.17 mg/g	
	HNO_3	Cr(III)		25.25 mg/g	
		Ni(II)		21.88 mg/g	
	Raw/drying	Cr(III)		13.5 mg/g	
		Ni(II)		12.03 mg/g	
Jackfruit seed	Drying	Malachite green dye	Freundlich	66 mg/g	[134]
	–	Eosin yellow dye	Langmuir	11.70 mg/g	[138]
Pineapple peel	Pyrolysis	Cr(VI)	Freundlich	7.44 mg/g	[143]
	1-butyl-3-methylimidazolium chloride	Methylene blue dye	Langmuir	101.94 mg/g	[141]
Pineapple crown	H_3PO_4/NaOH activation – pyrolysis	Methylene blue dye	–	99.48%	[139]
		Malachite green dye		98.94%	
Pineapple leaf	Carbonization	Cr(VI)	Langmuir–Freundlich	18.77 mg/g	[144]
Rambutan peel	KOH activation – CO_2 gasification	Remazol brilliant blue R dye	Langmuir	78.38%	[145]
	KOH activation	Acid yellow 17	Langmuir	215.05 mg/g	[147]
	–	Basic fuchsine dye	Freundlich	80%	[148]
Rambutan seed	KOH activation – CO_2 gasification	Malachite green dye	Freundlich	91.45%	[149]
Mangosteen peel	Microwaved assisted – K_2CO_3 activation	Methylene blue dye	Langmuir	379.65 mg/g	[151]
	KOH activation – CO_2 gasification	Remazol brilliant blue R	–	80.35%	[152]
	CaOH activation	Cr(VI)	Langmuir	2.46 mol/kg	[154]
	CaOH/NaOH activation	Cu(II)	Langmuir	1.48 mol/kg	[80]
		Fe(III)		1.20 mol/kg	
		Ni(II)		0.47 mol/kg	
		Cd(II)		0.35 mol/kg	

	Carbonization	Malachite green dye	Langmuir	31.45 mg/g	[157]
Pomegranate peel	–	Cr(II)	Langmuir	370.4 mg/g	[158]
	Fe(III) activation – carbonization	Cd(II)	Langmuir–Freundlich	22.72 mg/g	[162]
Pomegranate seed	–	Cr(VI)	–	3.31 mg/g	[160]
Melon peel/husk	Formaldehyde activation	Mixture of Methylene blue and Congo red dyes	Langmuir	26.42 mg/g	[165]
	H_2SO_4 activation	Cd(II)	Langmuir–Freundlich	73.5%	[167]
		Pb(II)		100%	
Melon seed	Microwave assisted – NaOH activation	Methylene blue dye	Langmuir	333.5 mg/g	[166]
		Acid blue dye		341.96 mg/g	
	Hydrochloric protonation	Methylene blue dye	Langmuir	489.8 mg/g	[168]
		Crystal violet dye		104.76 mg/g	
		Rhodamine B dye		86.6 mg/g	
Watermelon shell/rind	Xanthation activation	As(III)	Langmuir	99%	[172]
		As(V)		98%	
	ZnS nanoparticle activation	Pb(II)	Langmuir	106.4 mg/g	[82]
		Cd(II)		69.7 mg/g	
		Cu(II)		37.7 mg/g	
		Ni(II)		40.2 mg/g	
		Co(II)		32.8 mg/g	
	Pyrolysis	Methylene blue dye	Langmuir	95%	[176]
Durian shell/rind	Carbonization	Cu(II)	Langmuir	99.6%	[179]
	HCl activation	Zn(II)	Langmuir	36.73 mg/g	[180]
	NaOH activation	Neutral red	Langmuir	640.3 mg/g	[184]
		Malachite green dye		314.9 mg/g	
Grapefruit peel	H_2O_2 activation	Leather dye	Langmuir	80%	[81]
		Cr(VI)		100%	

where optimum adsorption capacity by 344.8 mg/g could be reached at pH 2. The performance of the sorbent enhanced linearly within 110 min of contact time before it became flat. This showed that the saturated condition of quaternary amine-modified orange peel-based sorbent has relatively long usage for the first adsorption cycle.

While some studies aforementioned reported acidic pH as optimum pH level, an investigation on the adsorption of Crystal violet and methylene dyes by orange peel derived sorbent reported a different point of view as its best removal efficiency happened at pH 6.5 within the first 30 min [113]. This work successfully achieved the removal percentage of both Crystal violet and methylene by 97.5% and 99.2%, respectively. Lam et al. [114] transformed orange peel waste into AC through microwave-assisted pyrolysis followed by chemical impregnation using alkali metal hydroxides for Malachite green dye removal. RSM adopting central composite design (CCD) was then implemented to maximize sorbent yield as well as adsorption efficiency. They found that the pyrolysis process having a heating rate of up to 112°C/min for 25 min could produce more energy-efficient than conventional longer heating by getting a sorbent yield of 87 wt.%. This satisfactory result may relate to the characteristics of AC made in this work showing high porous structure with fixed carbon up to 83 wt.% and Brunauer–Emmett–Teller (BET) surface area by 1,350 m^2/g.

For metal removal, the potentiality of orange peel as an effective biosorbent including for Cu(II) removal was also recognized [115]. Comprehensive analysis of variance (ANOVA) indicated that initial Cu(II) concentration and pH were significantly important. Based on the experimental work, they obtained optimum Cu(II) removal percentage by 94% at pH 5 with an initial metal concentration of 36 ppm, within 75 min of contact time. Then among Langmuir, Freundlich, Temkin, and Dubinin–Radushkevich isotherms applied, the Temkin equation seemed to have better suitability to the equilibrium data. Other studies also supported orange peel-based biosorbent for removing Cu(II) and informing that the adsorption kinetics seemed relatively fast within 60 min of contact time leading to Cu(II) maximum adsorption capacity of 63 mg/g [116].

7.2. Banana waste

Banana is among the most frequent fruits that have been widely studied as biosorbent materials exposing its potentiality to adsorb a wide variety of contaminants including dyes and metals [117]. Abu Bakar et al. [118] firmly confirmed that just 0.4 g banana peel-derived sorbent in 100 mL of Methylene blue solution could remove this unwanted pollutant from textile wastewater by more than 86.88%. Zhang et al. [119] then improvised banana peel becoming a green biochar/iron oxide composite involving $FeSO_4$ activation to adsorb Methylene blue and reported satisfying results. The maximum adsorption capacity of 862 mg/g could be achieved at initial dye concentration 500 mg/L, pH 6.1 and temperature 313 K. Its regeneration output showed relatively stable capacity even after five treatment cycles indicating that this modified biosorbent was economically reliable.

Further, Reactive Black 5 removal by chemically modified banana peel sorbent was found to be able to reach maximum monolayer adsorption capacity by 211.8 mg/g at acidic pH 3.0 [120]. In this study, the lack of adsorbed dye particles at basic pH could be related to the lower electrostatic occurrence regarding dye and chemical characteristics type. Oyekanmi et al. [121] researched different type of dye named Rhodamine B to be removed using acid-modified banana peel. Their experimental work revealed extremely acidic pH 2, sorbent dosage 0.2 g/L, and 60 min of contact time as optimum operating conditions following Langmuir isotherm as the best-fitted model. This study detected some acidic functional groups like carboxylic and hydroxyl groups on the surface of the biosorbent acting as a trigger for the successful removal of positively charged Rhodamine B dye molecules.

Bibaj et al. [44] proved that banana peel-derived sorbent was also effective for metal removal. They created biosorbent made from banana peel which was separately treated by hydrothermal and pyrolysis techniques to adsorb Ni(II) from an aqueous solution. The adsorption capacity of AC treated by pyrolysis and hydrothermal ranged from 415 to 488 m^2/g and from 315 to 320 m^2/g, respectively, both at the same optimum pH 6. It meant that pyrolysis outperformed hydrothermal activation by more than 15%. Li et al. [79] then made novel carbon foam from physically activated banana peel to simultaneously adsorb Cu(II), Pb(II), Cd(II) and Cr(VI) in an aqueous solution through a batch experiment. Within the first 5 min, they reported maximum sorption capacities of Cu(II), Pb(II), Cd(II) and Cr(VI) by 49.5, 45.6, 30.7 and 25.2 mg/g, subsequently. When they lengthened contact time up to 60 min, more than 98% of all metal ions could be successfully removed. In accordance with kinetic and equilibrium studies, pseudo-second-order kinetic and Langmuir isotherm was found as the most suitable models following thermodynamically endothermic and spontaneous mechanisms. Overall, compared to commercial AC, this modified carbon foam has a competitively higher removal efficiency.

Later on, Cr(IV) metal ion reduction was also successfully conducted using acidified banana peel-based sorbent by Ashraf et al. [122] and Ali et al. [123]. Ashraf et al. [122] made a comparison between the acid banana peel and organo-montmorillonite sorbents as a function of pH and initial pollutant concentration. They found that at pH 3, sorbent dosage 4 g/L, and temperature 20°C ± 1°C, the values of maximum Cr(IV) removal efficiency were 88% and 69% for acidified banana peel and organo-montmorillonite sorbents, respectively. This proved the superiority of biosorbent to its competitor. It was strengthened by the subsequent study through the investigation on biosorbent made from acrylonitrile grafted banana peel to remove the same metal type. In this work, biosorbent obtained higher Cr(IV) removal efficiency with relatively similar circumstances by pH 3 and sorbent dose 4 g/L yielding maximum removal efficiency of up to 96%. In general, both studies indicated that different activation modes on biomass may result in the different performance of biosorbent to adsorb the same pollutant.

7.3. Mango waste

Mango fruit contains nutritious elements including organic acids, sugars, flavor volatiles, phenolics, and lipid compounds [124]. To avoid a huge amount of its waste from overwhelming the environment, valorizing them into biosorbent is one of the palpable solutions. Some work proposed mango waste as effective sorbent material for Crystal violet dye depletion [125]. Shoukat et al. [126] then modified mango seed to become biocomposite for Crystal violet dye removal from textile effluent. They revealed promising results by 352.79 mg/g of adsorption capacity obtained at basic pH 8, sorbent dosage 0.05 g, contact time 30 min, and temperature 33°C. This work stated that both salt and surfactants pre-treatments for mango stone biocomposite have no significant effect on the adsorption of Crystal violet dye. In contrast, another study reported that the performance of carbonaceous biosorbent to remove eight types of acid dye was strongly influenced by ionic species in the solution related to the acidic/basic groups on the sorbent surface [127].

The influence of some physical and chemical properties such as bulk density, ash content, moisture content, iodine number, functional groups, surface morphology, and point of zero charge on the success of sorbate adsorption was also thoroughly observed by Jawad et al. [128]. They made mango peel-derived AC for Methylene blue removal and believed that those properties differently influenced the adsorption capacity of the sorbent which reached 277.8 mg/g. Literature also shows a wide variety of dyes that could be adsorbed by utilizing mango seed kernel including Malachite green dye which yielded maximum removal efficiency and adsorption capacity by 96% and 22.8 mg/g, respectively [129].

The use of mango cultivar peel and mango seed-based sorbents for metals removal also denotes positive output. Using these materials, Cr(VI) could be mostly removed from aqueous solutions at extremely acidic pH 1, initial Cr(VI) concentration 100 mg/L, and sorbent dosage 3 g/L [130]. Specifically, biosorbent made from mango seed outperformed mango cultivar by obtaining 100% while the other one only reached 97% of Cr(VI) removal efficiency. Overall, it indicates that both mango waste parts have the reliable potentiality for adsorbing Cr(VI) metal ions from aqueous media.

Moreover, different metals could result in different sorption capacities. A study utilized mango peel as biosorbent reached maximum adsorption capacity of Cr(III) and Cr(VI) at pH 5.0 and 7.0 by 98.039 and 66.666 mg/g, respectively [131]. However, Cu(II), Ni(II), and Zn(II) metal ions were reported to have lower sorption capacity using the same sorbent type by 46.92, 39.75 and 28.21 mg/g, respectively [132]. Therefore, proper activation has to be considered to equalize sorbent's ability in adsorbing targeted pollutants.

7.4. Jackfruit waste

Jackfruit (*Artocarpus heterophyllus* L.) is widely planted in Asia countries such as Thailand, Indonesia, Myanmar and India [117]. The valorization of its wastes including peel, seed, and leaf as biosorbent would not only increase its economic value, but also reduce disposal management costs. The utilization of dried jackfruit leaf to adsorb Malachite green dye showed a promising outcome [133]. In this work, the characterization analysis indicated that jackfruit leaf powder has a large number of pores, spaces, and empty sites to adequately attach pollutant particles. Despite strong adsorption bonding between sorbent and pollutant due to the presence of functional groups such as –OH, –NH and C=O on the sorbent surface, around 80% of the attached Malachite green dye could still be released in the desorption process leading to more economical aspects. Kooh et al. [134] then used jackfruit seed to remove Malachite green dye via batch adsorption experiment and found that the adsorption process was a high reliance on the solution pH and less dependence on ionic strength.

Further, carbonized jackfruit peels and seeds derived-sorbent could also be employed to adsorb copper metal ions [135]. This study claimed an excellent adsorption capacity of up to 99.84% that could be achieved at temperature 45°C and neutral pH 7 within 24 h with the initial concentration of copper solution 100 ppm. The investigation of jackfruit leaf derived-sorbent modified by chemical impregnation using KOH and H_3PO_4 to remove Cr(VI) from wastewater was also reported [136]. They declared that H_3PO_4 impregnation gave a better performance to biosorbent than KOH solution by reaching higher metal removal. Eventually, another study observed jackfruit peel surface modification using four chemicals separately named ethylenediaminetetraacetic acid (EDTA), sodium dodecyl sulfate (SDS), NaOH, and HNO_3 [137]. They proved that, compared to its raw version, jackfruit-based sorbent modified by those chemicals experienced multiplication in terms of its adsorption capacity on Cr(III) and Ni(II) ions due to changes in physicochemical characteristics on the biomass such as the number of the active binding site and ion-exchange properties.

7.5. Pineapple waste

The wastes of pineapple (*Ananas comosus*) such as peel, core, crown, and leaf can be utilized as biosorbent material. Raw pineapple peel, for example, is powerful to remove an anionic Eosin yellow dye by gaining an adsorption capacity value of 11.76 mg/g [138]. Thus, Selvanathan and Subki [139] studied carbonized pineapple crown, core, and peel to repeal Methylene blue and Malachite green dyes from the solution. Those wastes were separately prepared by chemical activation using H_3PO_4 and NaOH. The results indicated that AC derived from pineapple crown was superior to others by reaching Methylene blue and Malachite green dyes removal efficiency of up to 99.48% and 98.94%, respectively. Different chemicals are assumed to have a different effects on biosorbent surface characteristics. Therefore, composite hydrogels prepared from pineapple peel cellulose and sepia ink involving other chemical acetylation types named ionic liquid 1-butyl-3-methylimidazolium chloride [140]. The work informed that sepia ink could improve adsorption capacity significantly from 53.72 to 138.25 mg/g when its portion in the hydrogels was 10%. They then continued investigating pineapple peel in the form of cellulose/magnetic diatomite hydrogels using 1-butyl-3-methylimidazolium chloride to adsorb Methylene blue from liquid solution [141]. After characterization, they indicated that the existence of magnetic diatomic increased the swelling ability and adsorption capacity. The equilibrium adsorption could be obtained in the first 30 min following pseudo-second-order kinetic and Langmuir isotherm models with a maximum adsorption capacity of 101.94 mg/g.

Further, other study valorised pineapple peel as biosorbent using water, alkali, bleaching, and bleaching-alkali activation in separate modes followed by hydrogels fabrication [142]. This led to 114.94, 77.52, 138.89, and 75.19 mg/g of adsorption capacity for those solutions, respectively. This work also demonstrated that the adsorption process followed the pseudo-second-order model by reaching coefficient correlation close to 1 implying a chemisorption process through sharing or exchange of electrons between Congo red cations and sorbent functional groups.

Regarding metal depletion, an investigation on biochar made from pineapple peel for Cr(VI) removal revealed the influence of pyrolytic temperature and contact time on sorbent capability [143]. The adsorption capacity of 7.44 mg/g was reached at pyrolytic temperature 750°C and initial metal concentration 10 mg/L. Moreover, this biochar needed 18 h to be in the adsorption equilibrium where its adsorption kinetics fit well the second-order kinetic equation. The performance of this biochar may be caused by the formation of an inner complex between hydroxyl groups on sorbent and pyrene leading to metal ion inhibition. However, Ponou et al. [144] could get a better output of pineapple leaf sorbent carbonized at lower temperature by around 450°C. It successfully removed Cr(VI) with removal efficiency and adsorption capacity of 90.1% and 18.77 mg/g, respectively. This might relate to more functional groups available in the pineapple leaf than peel. Other than that, better biosorbent characteristics could be formed at certain pyrolytic temperatures. This study also recognized the chemisorption mechanism by following a pseudo-second-order kinetic model.

7.6. Rambutan waste

Rambutan has a unique outlook and nutritious contents. Apart from its consumable part, rambutan generates by-products such as peels and seeds which are usually disposed making them considerable as low cost sorbent biomass. Ahmad and Alrozi [145] used rambutan peel-derived sorbent activated by KOH and CO_2 gasification for Remazol brilliant blue R dye removal. By applying ANOVA, they found that AC prepared at temperature 789°C for 1.8 h resulted in 78.38% of dye removal efficiency. Alrozi et al. [146] then observed rambutan peel derived sorbent for removing the same dye and reported an increasing adsorption uptake with the increase in initial concentration and contact time where adsorption capacity of 112.69 mg/g could be obtained at pH 4.

These promising results inspired other studies to investigate other types of dyes and modification methods namely microwave heating and KOH impregnation that were applied to rambutan peel before it was used as biosorbent to adsorb acid yellow dye [147]. With a maximum adsorption capacity of 215.05 mg/g, they claimed this high adsorption capacity occurred at both low and high initial dye concentrations. Further, this biosorbent could also remove fuchsine dye in raw textile wastewater by reaching 80% of removal efficiency where maximum adsorption capacity 108.969 mg/g could be achieved around 3 h of contact time [148].

Other than its peel, rambutan seed also showed a comparable outcome. Ahmad et al. [149] activated the seed using KOH impregnation and CO_2 gasification for Malachite green dye adsorption. The experimental works were considered based on the controlled parameters during the adsorption processes such as activation temperature, activation time, and chemical impregnation ratio (IR) (KOH: char by weight). By employing modeling CCD and ANOVA, they found activation temperature 802°C and activation time 60 min as optimum conditions resulting in maximum dye removal of up to 91.45%.

Regarding metal removal, rambutan peel-derived-biochar processed under different pyrolytic temperatures ranging from 300°C to 700°C has shown a promising view [150]. A solution containing Cu(II) metal could be treated well using this biochar especially biochar which was pyrolyzed at a temperature of 600°C.

7.7. Mangosteen waste

As one of the seasonal tropical fruits, mangosteen is well-known for its delicate texture and exotic taste. Despite its limited production, valorizing its wastes can be useful to find the alternative environmentally friendly sorbent material, then some studies were conducted in order to evaluate the performance of both raw and modified mangosteen wastes-derived sorbent.

To begin with, the efficacy of microwave-assisted K_2CO_3 activation on mangosteen peel to repeal Methylene blue from aqueous solution was notified [151]. Based on their experimental results, the maximum monolayer adsorption capacity obtained was 379.63 mg/g with a carbon yield of 80.95% indicating its positive tendency as a reliable biosorbent. Ahmad and Alrozi [152] tried different modification types using KOH impregnation and CO_2 gasification to make AC from mangosteen peel for Remazol brilliant blue R removal. They found that the effects of the three selected variables namely; CO_2 activation temperature and time, and KOH impregnation ratio on dye percentage removal were noticeable. Specifically, at temperature 828°C, activation time 1 h, and impregnation ratio 3.0 resulted in 80.35% of targeted dye removal. The effectiveness of mangosteen peel sorbent modified by ultrasound integration on Reactive Black 5 dye removal from solution was also competitive by having an incredible dye removal efficiency of more than 95% even at neutral pH [153].

Huang et al. [154] then continued using mangosteen peel-based sorbent to remove Cr(VI) metal ions. It was initially activated using a $Ca(OH)_2$ solution. They informed that biosorbent worked better at an extremely acidic solution where pH 1, 2, 3, and 4 resulted in 2.46, 2.44, 1.99, and 2.14 mol/kg of adsorption capacity, respectively. Interestingly, the adsorption mechanism analyzed matched an esterification reaction coupled with the reduction of Cr(VI) to Cr(III). Another study created biosorbent gel made from mangosteen pericarp activated by saponification process using the alkaline solution, $Ca(OH)_2$ and NaOH to eliminate selected metals contained in the solution [80]. After analysis, the selectivity order for heavy metal ions found to be Fe(III) > Pb(II) > Cu(II) > Cd(II) > Zn(II) > Ni(II). In addition, maximum adsorption capacity of 1.48, 1.20, 0.47, 0.35 mol/kg were also revealed for Cu(II), Fe(III), Ni(II), Cd(II), respectively. Finally, Zein et al. [155]

suggested mangosteen shell-derived sorbent to remove low concentrations of Pb, Zn, and Co from the aqueous solution which could reach metal removal rates of up to 90% within 60 min of contact time and pH up to 5.

7.8. Pomegranate waste

Pomegranate fruit (*Punica granatum* L.) has considerable by-products including pomegranate peel and seed [117]. Therefore, several studies attempted some research in order to valorize them. For instance, harmful Malachite green dye could be eliminated using carbonized pomegranate peel [156]. At pH 6, initial dye concentration 30 mg/ L, higher temperature 50°C, and contact time 90 min, the maximum adsorption capacity of 31.45 mg/g could be achieved even with 0.1 g of this biosorbent while its adsorption model followed Langmuir isotherm and pseudo-first-order kinetics [157]. Further, Salam et al. [158] conducted the eradication of Cr(VI) in batch mode and, after evaluation, they informed a remarkable maximum removal of 100% at pH 2, temperature 313 K, and contact time 3 min. Other research then used raw pomegranate peel to remove Cu(II) ions and figured that the highest adsorption capacity occurred at pH 5.8, temperature 313 K, and contact time 120 min [159].

Regarding the seed, Ghaneian et al. [160] examined it to detach Cr(VI) from contaminated water and found that adsorption percentage increased with the increase in biosorbent dosage where the adsorption capacity was determined better at pH 2 with an initial metal concentration of 10 mg/L. Ali et al. [161] showed the performance of pomegranate peel to simultaneously adsorb both metal and dye contaminants namely copper ions and C.I. reactive yellow 145. By modifying pomegranate peel into ball-milled sorbent. They believed that the maximum removal capacity of this biosorbent for reactive yellow and copper ions were 209.7103 and 103 mg/g, respectively. Meanwhile, Abedi et al. [162] studied the effectiveness of Fe(III) modified pomegranate peel carbon for Cd(II) removal. They discovered that the adsorption capacity of biosorbent obtained a good degree by 22.72 mg/g following the pseudo-second-order kinetic model.

7.9. Melon waste

Melon fruit belonging to the *Cucurbitaceae* family provides high vitamins and antioxidants, and it was estimated to have millions of tons of production capacity worldwide [163]. As a consequence, its waste is an overwhelming environment. Then like other investigations of plant-based biosorbents, Djelloul and Hamdaoui [164] deployed biosorbent made from melon peel for Methylene blue dye removal from aqueous solutions. In their report, a maximum monolayer adsorption capacity of 333 mg/g was stated using the Langmuir isotherm formula indicating a promising pattern for future research.

It is firmly argued that raw industrial wastewater may have multiple dye species, so characteristics of a particular dye in the water could be affected by the presence of the others. Then the adsorption of Methylene blue in both single and ternary dye systems consisting of three different prominent dyes, namely Methylene blue, Congo red and methyl orange using formaldehyde-treated melon husk was investigated to prove it [165]. The experimental work resulted in dye adsorption equilibria which were attained after 60 min for a single dye system and 90 min for the ternary dye system. In addition, the adsorption kinetics was well described by the pseudo-second-order model while the equilibrium adsorption data was analyzed by six different isotherm models indicating the Langmuir formula as the best fitted one. Melon seed hull-derived AC modified through microwave-assisted NaOH activation could actively adsorb Methylene blue and Acid blue 15 by 333.50 and 341.96 mg/g, respectively, as reported by Foo et al. [166]. They also informed typical data such as BET surface area, Langmuir surface area, and total pore volume by 1,187 m^2/g, 1,804 m^2/g, and 0.68 cm^3/g, respectively.

Eventually, metal contaminants like Cd(II) and Pb(II) can also be brought into the pores of biosorbent made from melon peel activated by H_2SO_4 [167]. Excellently, it obtained complete removal efficiency by 100% with lower dependency on the prominent parameters.

7.10. Watermelon waste

Watermelon is a watery fruit containing a high percentage of water element protected by a sturdy shell which can be utilized as an alternative low-cost biosorbent. Literature shows that both dyes and metals could be successfully separated from contaminated water and wastewater using its waste. To begin with, three different dyes could be removed simultaneously from the aqueous solution using the protonated watermelon rind-derived biosorbent [168]. In this work, hydrochloric acid was used during the protonation process resulting in a rapid equilibrium level within 30 min with maximum adsorption capacity of 489.8, 104.76, and 86.6 mg/g, for Methylene blue, Crystal violet, and Rhodamine B, respectively. Desorption analysis was separately performed using HCl, CH_3COOH, H_2O, and NaOH proving that different chemicals would give different results on desorption values. They then expanded the study by investigating the same biosorbent type for Methylene blue removal in batch and continuous column modes [169]. It was found that both systems were reliable. While equilibrium data followed both Langmuir and Temkin models, kinetic data indicated pseudo-second-order kinetic model and spontaneous exothermic thermodynamic process.

Furthermore, Chigbundu et al. [170] worked on the usage of watermelon rind derived biosorbent for cationic basic red 2 and anionic orange dyes, respectively. The equilibrium data were well represented by the extended Langmuir isotherm equation where the adsorption capacity of basic red 2 and orange dyes was to be 125 and 27 mg/g, respectively. This significant gap of adsorption capacity between these two dyes may be related to the physical–chemical characteristics of the dye itself.

Watermelon biomass also has drawn positive reviews for metal removal. Banerjee et al. [171] could reach the adsorption capacity of Cu(II) by 111.1 mg/g using a biosorbent made from watermelon rind. They suggested pseudo-second-order kinetics supported by a particle diffusion process. In order to increase metal adsorption, this study advised a few steps such as decreasing both initial dye concentration and particle size, increasing pH up to 8, and increasing temperature and agitation speed. Moreover, Shakoor et al. [172] reported the success of both As(V) and As(III) metal ions eradication from aqueous solutions. They implemented separately raw, xanthated modified, and citric acid-modified watermelon rind-based biosorbent. Among others, the potentiality of xanthated watermelon rind was then confirmed better with relatively higher As(V) and As(III) removal efficiency. When the initial concentration of each metal was 4 mg/L with sorbent dosage 1 g/L, for example, they could get the adsorption percentage by 99% at pH 8.2 for As(III) and 98% at pH 4.6 for As(V). In addition, compared to its raw version, xanthated watermelon rind showed up to 49% greater As(III) and As(V) removal efficiency indicating the idiosyncratic leverage of chosen modification. Another study produced an activated biosorbent made from watermelon rind by involving ZnS nanoparticles and reported satisfactory loading capacity of the biosorbent for adsorbing heavy metals namely Pb(II), Cd(II), Cu(II), Ni(II) and Co(II) [82].

Furthermore, Li et al. [173] treated thallium (Tl(I))-containing wastewater using watermelon rind derived-porous biochar pyrolyzed at temperature 500°C. They then got the maximum Tl(I) adsorption capacity by 178.4 mg/g. After conducting characterization, it was depicted that K- and Cl-rich primary biochar has a promising regeneration of amphibious ion exchange resins for both adsorption and desorption processes. Finally, Lakshmipathy et al. [174] used raw watermelon rind sorbent to remove both Ni(II) and Co(II) ions from aqueous solution conducted in batch mode and obtained maximum adsorption capacity by 35.3 and 23.3 mg/g for Ni(II) and Co(II), respectively. Specifically, they proposed the rivalry theory between those metals related to the existence of the ions extraction process affecting the amount of metal ions adsorbed onto sorbent pores.

7.11. Durian waste

As one of the widely consumed fruits in South East Asia region such as Malaysia, Thailand, and Indonesia, the production number of durian follows its high demand leading to the accumulation of durian waste such as durian peel which is also known as durian husk [175]. A magnetic biochar made from durian rind in the presence of iron oxide was reported being able to adsorb Congo red dye from polluted solution [176]. Good performance was reported by reaching 98% of dye removal while adsorption capacity 87.32 mg/g scattered on 820 m^2/g of biosorbent total surface area.

Kurniawan et al. [177] removed most Cr(VI) content from synthetic wastewater by durian shell based biosorbent and successfully got 117 mg/g of maximum adsorption capacity. Looking at its capability on metal adsorption, other study then proved it by trying to simultaneously remove Crystal violet and Cr(VI) ions from aqueous solution by rarasaponin–bentonite-activated biochar made from durian shells composite [178]. According to this investigation, there was a contradictory preferred temperature between those pollutants. While Crystal violet adsorption performed better at higher temperatures, Cr(VI) was better at lower temperature. Therefore, the characteristics of each targeted pollutant and choosing suitable operating condition are part of the influential aspects in the success of pollutant removal efficiency. Bach et al. [179] added to the literature that AC from this fruit shell was able to put Cu(II) at bay with excellent removal by 99.6%. Moreover, they also found this biosorbent was cost effective due to having great regeneration up to five times with relatively stable output in each cycle.

Hence Ngabura et al. [180] informed their spectroscopic analysis on HCl activation biosorbent by describing that the predominant contributor for adsorption process on the sorbent surface was the existence of hydroxyl, carbonyl, carboxyl and amides groups. Their work was then continued to investigate Pb(II) sorption from synthetic solution by employing RSM simulation [181]. After conducting first experiment set at pH up to 7.0, biosorbent dosage 0.39 g, contact time 60 min, and initial Pb(II) concentration 100 mg/L, they achieved maximum adsorption capacity by 14.6 mg/g. However, in the second experiment set at temperature 41.27°C, pH 8.95, and initial metal concentration 99.96 mg/L, the maximum adsorption capacity decreased to become 9.67 mg/g. This means that operating conditions especially pH hold an important role to some extent.

7.12. Grapefruit waste

Grapefruit is relatively popular fruit as it can be cultivated in both tropical and subtropical regions. Even though its peel is a consumable part, the curiosity about its efficacy as sorbent materials has led to several investigations. This was triggered by its rich carboxyl and hydrocxyl functional groups contents notable for binding targeted pollutant in aqueous solution [117]. Therefore, a variety of dyes and metal ions were successfully eliminated by grapefruit peel both in its raw and modified version. Najafinejad et al. [182] implemented Fe_3O_4-polythiophen-Ag magnetic nanocatalyst derived from grapefruit peel to adsorb both methyl orange and Methylene blue dyes. They concluded that reaction rate in the presence of catalyst during activation process was very considerable as both dyes could be reduced significantly even within 1 min of contact time. It was worthy to reveal the incredible recovery of this biosorbent as it could be stable even after the eighth adsorption cycle. The next discussion is magnetic composite made from grapefruit peel activated using Fe_3O_4 solution to simultaneously remove Congo red, humic acid, and phosphate from aqueous solutions [183]. This work disclosed that the spreading of Fe_3O_4 particles on the biosorbent surface has a strong correlation to the higher level of magnetism characteristics leading to 92.88%, 47.45%, and 99.02% of Congo red, humic acid, and phosphate removal efficiency, respectively. Additionally, different from mostly related studies about biosorption, this work found that its adsorption profile has no dependence on solution pH.

Furthermore, NaOH modified grapefruit peel used to expel synthetic dyes out from contaminated solution attained favourable maximum equilibrium quantities for Neutral red and Malachite green dyes by 640.3 mg/g and 314.9 mg/g at 298 K, respectively [184]. The adsorption equilibrium of Neutral red fitted well with both the Langmuir and Koble-Corrigan models while Malachite green was more suitable to Koble-Corrigan and Dubinin-Radushkevich models. They also proposed binary system as more favourable mode for both dyes as it could exhibit more competitive adsorption than single system.

Other complacent output reported by Rosales et al. [81] declared that leather dyes and Cr(VI) content in wastewater could be depleted using grape peel-derived sorbent modified by H_2O_2 activation. The sorbent enabled to reach the level of removal efficiency by 80% and 100% for leather dyes and Cr(VI), respectively. Eventually, Zhang et al. [185] created a modified grapefruit peel based sorbent named biochar/pectin/alginate hydrogel beads (BPA) and reported that it brilliantly worked well to eliminate Cu(II) from aqueous solution in batch modes. They found that around 80.6 mg/g of biosorbent adsorption capacity could be reached at optimum pH 6 with spontaneous feasibility and endothermic nature of the adsorption behaviour.

8. Conclusions, challenges, and future outlook

8.1. Conclusions

In this review article, assorted biosorbents made from various fruits waste to remove dyes and heavy metals have been presented. Comprehensive literature related to the characteristics, usages, and effects of those dyes and heavy metals on human health as well as various existing technologies for their removal are also explained. This phenomena relates to a growing industrialisation that has generated more and more dye and heavy metal pollutants contained in water and wastewater stream threatening human health and environmental sustainability. Hence, the existence of environmentally friendly wastewater treatment technology is one of critical solutions. For this reason, this article reveals the advantages and disadvantages of numerous treatment methods such as membrane filtration, coagulation/flocculation, biological technique, adsorption, and hybrid methods to some extent. This will lead to a better perception of each method in terms of its efficiency and effectiveness on dyes and heavy metals eradication. Eventually, biosorption process using fruit waste based-sorbents seems still well considered due to its high renewability, lower cost, and abundant choices and availability. To conclude, implementing modification techniques on biosorbent materials as well as finding optimum conditions could noticeably enhance the adsorption capacity and pollutant removal efficiency.

8.2. Challenges and future outlook

Based on the literature study, research articles related to the usage of fruit waste as main biosorbent materials for adsorbing metals and dyes mainly focused on the technical and performance aspects. However, there are some particular limitations regarding cost effectiveness analysis, especially when it is compared to the available commercial sorbents or biosorbent derived from other organic materials. By considering this point of view, it will be beneficial for upgrading them into wide scale application in real industrial wastewater treatment plant. Moreover, several inconsistencies between the experimental and theoretical or predicted uptake of sorbent capacity still could be found resulting in less ideal condition of biosorbent surface for its wide scale usage. Another challenge is how to perform low cost and safe regeneration process as it can lead to durable sorbent character along with sustainable disposable modes. More research studies then should be conducted for the comprehensive application of novel materials in wastewater treatment as it can be a reliable alternative to readily available conventional sorbents [185].

Furthermore, in most studies, the removal efficiency of sorbents for both dyes and heavy metal significantly increased after modification. Therefore, more functionalization types on the conventional and novel biomaterials are worth to be investigated for future perspectives. It is also worth to conduct more comparative research among modification ways, operating conditions, performance and properties of the available and novel sorbents derived from biomass. Eventually, it is critical to employ both simulated and relevant raw industrial wastewaters as solution sample for better comparative perception as some particular pollutants existing in the industrial wastewater may have significant effect on the effectiveness of modified biosorbent when they are contacted with each other in the real conditions.

Symbols

q_e — Amount of sorbate adsorbed per unit of sorbent at equilibrium time, mg/g
C_e — Equilibrium sorbate concentration in the solution, mg/L
k_f — Isotherm constant
n — The adsorption intensity
q_m — Maximum adsorption capacity, mg/g
K_L — Langmuir constant
β — Constant related to adsorption energy
ε — The Polanyi potential
R — Gas constant, 8.314 J/mol K
T — Absolute temperature, K
K_{id} — Rate constant of intraparticle diffusion, mg/g min$^{0.5}$
$t^{0.5}$ — Square root of time, min

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