Chemosphere 338 (2023) 139477

Contents lists available at ScienceDirect

Chemosphere

journal homepage: www.elsevier.com/locate/chemosphere

Biogenic adsorbents for removal of drugs and dyes: A comprehensive review on properties, modification and applications

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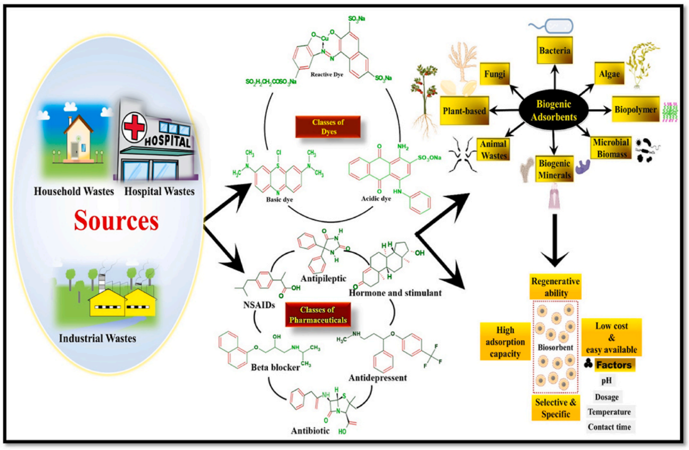
HIGHLIGHTS

- Review on biogenic materials for removal of drugs and dyes from wastewater.
- Adsorbents derived from plants, animals, microorganisms, algae and biopolymers.
- Mechanical, thermal and chemical modifications can enhance adsorption properties.
- Biogenic hybrid composites have enhanced adsorption properties.
- Reusability, limitations and future prospects of biogenic adsorbents are discussed.

GRAPHICAL ABSTRACT



ARTICLE INFO

Handling Editor: Y Yeomin Yoon

Keywords:
Biogenic materials
Biopolymers
Adsorption
Wastewater
Drugs
Dyes
Plant-based adsorbents
Biosorbents
Biomass

ABSTRACT

This comprehensive review explores the potential and versatility of biogenic materials as sustainable and environmentally benign alternatives to conventional adsorbents for the removal of drugs and dyes. Biogenic adsorbents derived from plants, animals, microorganisms, algae and biopolymers have bioactive compounds that interact with functional groups of pollutants, resulting in their binding with the sorbent. These materials can be modified mechanically, thermally and chemically to enhance their adsorption properties. Biogenic hybrid composites, which integrate the characteristics of more than one material, have also been fabricated. Additionally, microorganisms and algae are analyzed for their ability to uptake pollutants. Various influential factors that contribute to the adsorption process are also discussed. The challenge, limitations and future prospects for research are reviewed and bridging gap between large scale application and laboratory scale. This comprehensive review, involves a combination of various biogenic adsorbents, going beyond the existing literature where typically only specific adsorbents are reported. The review also covers the isotherms, kinetics, and

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https://doi.org/10.1016/j.chemosphere.2023.139477
Received 21 May 2023; Received in revised form 8 July 2023; Accepted 10 July 2023
Available online 11 July 2023

desorption studies of biogenic adsorbents, providing an improved framework for their effective use in removing pharmaceuticals and dyes from wastewater.

1. Introduction

The escalating threat of climate change reached a critical level, demanding immediate attention due to its profound impacts on the Earth's ecosystem (Raza et al., 2023b,c). One of the most pressing environmental issues resulting from industrial development is water contamination, which poses significant risks to both human health and ecosystems (Jaleh et al., 2023; Keyikoğlu et al., 2022, 2023). The detrimental effects of industrial and economic growth led to widespread environmental deterioration, particularly in the form of water contamination caused by the discharge of drugs and dyes. Sectors such as textiles, cosmetics, and pharmaceutical industries contribute significantly to this pollution. However, a promising and sustainable solution lies in the exploration of adsorbents derived from greener and renewable resources. By utilizing these eco-friendly materials, we can address the sources of contamination and implement effective remediation measures that will protect the environment and promote a healthier future (Raza et al., 2022, 2023b).

As the need for sustainable development and environmental protection becomes increasingly urgent, researchers and environmentalists are exploring the potential of biogenic materials as a substitute for synthetic adsorbents in various applications, including the removal of drugs and dyes from wastewater. Biogenic materials are not only naturally abundant but show also important physiochemical properties, surface morphology, structure and functionalities (Kumar et al., 2020). They are also efficient for water desalination (Raza et al., 2023a). The continuous discharge of dyes, drugs and other organic pollutants is a global concern, posing a significant threat to public health and the environment (Yadav et al., 2021). The depletion and contamination of aquifers by toxic substances needs urgent action to raise awareness of water-related issues caused by the continuously increasing population (Amin et al., 2014). Untreated dyes from textile industries cause environmental contamination, disrupting the overall food chain (Ahmad et al., 2017). Water quality is inadequately regulated and managed, leading to unwanted materials and toxins in groundwater and drinking water (Yaqoob et al., 2020). Dyes are complex organic compounds that are usually soluble in water and oil, and contain unsaturated compounds known as chromophores, which give them their vibrant and often fluorescent colors (Hunger, 2007). There are three major types of dyes used in textile industries: acid, basic, and reactive dyes (Fig. 1). Acid dyes are water-soluble and have a negative charge on their molecule. They require an acidic environment to bond with the fabric fibers. These dyes produce bright and vibrant colors, and the dyeing process can be carried out at a relatively low temperature. Basic dyes are water-soluble cationic dyes, meaning they have a positive charge on their molecule. They are used usually applied to the fabrics in alkaline conditions. The color produced by basic dyes is typically brighter and more vivid than acid dyes. Reactive dyes are water-soluble that react chemically with the fiber molecules to form a covalent bond, resulting in excellent color fastness. They require a neutral to alkaline environment for the dyeing process. The colors produced by reactive dyes are generally bright and long-lasting.

Worldwide industries use more than 10,000 different dyes, with more than 700,000 tons of various dyes and pigments produced annually. Approximately 10–15% of dyes are discharged into water bodies during manufacturing or other applications. The tremendous rise of dyes release in aquifers has undesirable effects on water quality due to their toxicity, resistance to degradation and carcinogenicity (Chequer et al., 2013; Yadav et al., 2021). Textile industries are responsible for discharging 200,000 tons of dyes annually during dyeing, making the water from these industries the most polluted regarding quantity and composition (Chequer et al., 2013). Dyes are mutagenic and toxic, preventing light penetration into water sources, which reduces photosynthesis and leads to undesirable effects on fauna and flora.

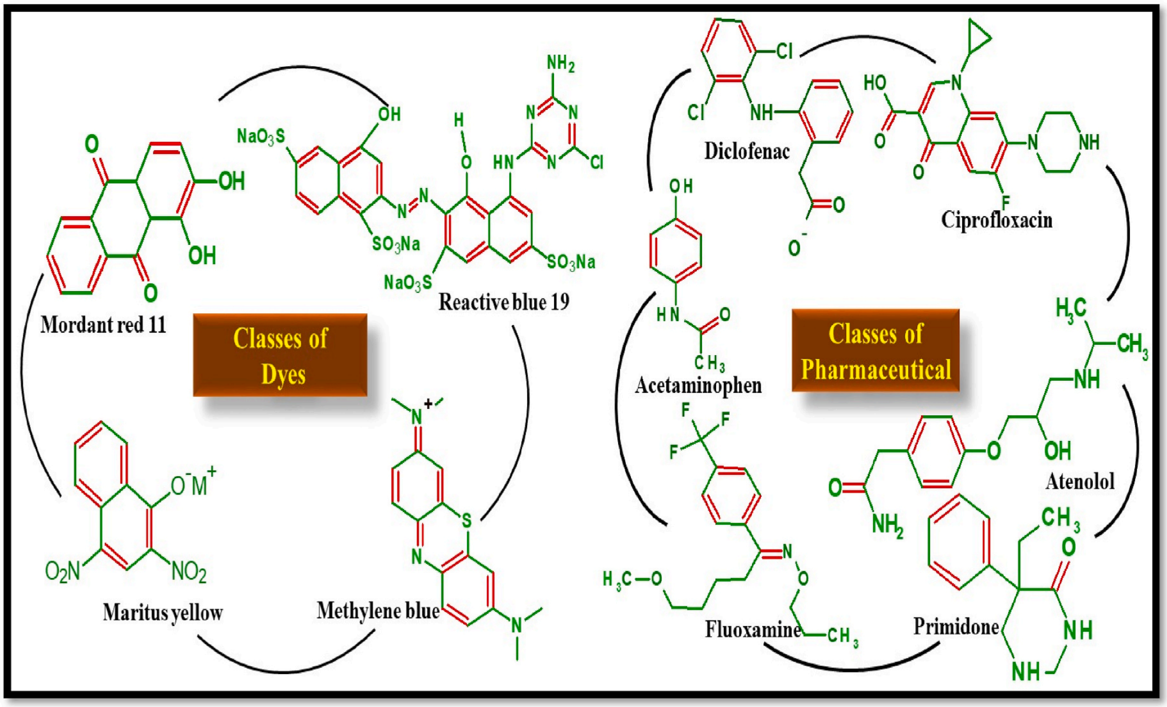

Fig. 1. Classes of drugs and dyes.

In addition, emerging active pharmaceutical ingredients (APIs) are constantly appearing in groundwater, freshwater sources and urban water sources, having adverse effects on the environment and human health (Raza et al., 2023a; Yadav et al., 2021). Non-steroidal anti-inflammatory drugs, antibiotics, hormones, antiepileptic drugs, antihistamines, blood lipids lowering agents, beta-blockers and steroid antidepressants have been recognized in water sources (Patel et al., 2019; Zare et al., 2022). Some examples of different dyes and drugs are shown in Fig. 1. The global excessive use and inappropriate disposal of pharmaceutical products, including nonsteroidal anti-inflammatory drugs (NSAIDs) pose a significant challenge, since they are often persistent and cause ecological toxicity (Yang et al., 2022)().

Several chemical and physical approaches, including coagulation, reverse osmosis, photocatalytic or microbial degradation, membrane separation, adsorption, and advanced oxidation processes, offer potential for the removal of dyes and drugs from aqueous environments (Mu and Wang, 2016). Advanced oxidation techniques, such as ozonation, Fenton and ultraviolet (UV) treatment are employed to specifically remove certain compounds. However, it is important to note that these processes can be expensive and generate toxic intermediates during complete oxidation, which may also exhibit toxicity (Kyzas et al., 2015).

Adsorption is one of the most effective methods for removing pollutants from various sources, such as textiles, cosmetics, plastics and pharmaceutical industries, which all end up in water sources without proper treatment. The primary benefit of this method involves simplicity, higher selectivity and availability of adsorbent materials (Mosoarca et al., 2022; Sartape et al., 2017). However, it is important to note that the efficiency of adsorption can vary depending on the specific characteristics of the adsorbent and the adsorbate. In the case of dyes and pharmaceutical compounds, adsorption can be particularly advantageous. Drugs, being organic compounds, can be effectively adsorbed onto certain adsorbents, enabling their removal from wastewater or other matrices. Dyes, commonly used in industries, can cause environmental concerns if not properly treated. Adsorption provides an efficient means of removing dyes from wastewater streams, leading to reduced environmental pollution.

In addition to organic compounds, heavy metals such as mercury, cadmium, and lead pose significant health risks due to their toxic persistence (Zaimee et al., 2021). Adsorption is a commonly employed method for removing heavy metals from contaminated water sources (Budiana et al., 2021; Khera et al., 2020; Neolaka et al., 2021, 2023; Zaimee et al., 2021). For example, of telescope snail and powder of mangrove crab shell as adsorbents for Pb^{2+} metal ion removal from aqueous samples (Darmokoesoemo et al., 2016a). Horn snail and mud crab shells were used for the removal of copper (Darmokoesoemo et al., 2016b). An adsorbent made from a mixture of bagasse-bentonite was used for removing Cd^{2+} (Kuncoro et al., 2018b) and Pb^{2+} (Kuncoro et al., 2018a,c) (bagasse was also efficient for adsorption of ciprofloxacin (Peñafiel et al., 2021) and diclofenac sodium (Antunes et al., 2012) from water). The selective adsorption capacity of ion imprinting polymer derived from kesambi wood and graphene oxide (GO) was investigated for Cr(VI), Pb(II), Mn(II), and Cd(II) in the same medium (Neolaka et al., 2020a,b). Algae waste-bentonite adsorbent was also used for the removal of heavy metals (Kuncoro et al., 2018a–c) Khera et al. investigated the promising adsorption capacity of *Archontophoenix alexandrae* against metals ions in single, bi and tri metallic systems (Khera et al., 2020). Takari sand-derived silica@mercapto hybrid composite was successfully used for the adsorption of Cu and Pb ions at pH 5 (Naat et al., 2021). Ngueagni et al. conducted a study on the fabrication and modification of hydroxyapatite derived from cattle horn cores for the purpose of removing Pb and Cd ions from water samples (Ngueagni et al., 2022).

While adsorption can be utilized for the removal of heavy metals, drugs, and dyes, it is important to acknowledge that each group may present specific characteristics and challenges that require tailored approaches for optimal results.

The exploitation of a large quantity of naturally occurring plant-based adsorbents leads to the complete removal of organic pollutants because plants majorly contain cellulose, lignin, hemicellulose and many other pigments, which offer multiple adsorption sites for the retention of dyes and drugs (Mosoarca et al., 2022). The binding of an effective adsorbent to a diverse range of aquatic pollutants is determined by favorable surface physicochemical properties of the adsorbent material. The enhanced variety of pharmaceutical and textile pollutants requires faster kinetics, greater adsorption capacity at higher concentrations and different pH (Nayak and Bhushan, 2021). Furthermore, the use of greener bio-inspired materials tends to revolutionize science by mitigating environmental pollution and clean green energy production (Kumar et al., 2020).

2. Biogenic adsorbents and their chemical nature

Biogenic adsorbents, including plant-based materials (e.g., peat, straw, sawdust), microorganisms (e.g., algae, bacteria) and biogenic minerals (e.g., clay, zeolites), are derived from natural sources and can be used for environmental bioremediation. They offer several advantages over synthetic adsorbents, such as low cost, biodegradability, and the ability to remove a wide range of emerging pharmaceutical active compounds and hazardous dyes from water bodies (Kumari et al., 2019; Natarajan et al., 2022). Additionally, biochars can be produced from various types of biomass through pyrolysis and show properties related to their feedstock. These stable carbonaceous derivatives have a number of active functional groups on the outer surface (Mohamed and Mahmoud, 2020b). Biogenic adsorbents are sustainable, versatile and environmentally friendly materials that have proven to be highly effective in removing pollutants, surpassing even some synthetic adsorbents in their performance (Kumar et al., 2020). These bio-sourced materials include plant-based, animal-based, living organism-based biomass, biopolymers, biogenic minerals and microorganisms (Karimi-Maleh et al., 2022; Malik et al., 2015; Yadav et al., 2021). Waste plant residue is a good candidate for developing ideal adsorbents. Various modifications are being carried out, either physical or chemical, to attain the desirable decontamination efficiency, by π-π interaction, ion exchange, electrostatic interaction or attraction, hydrogen bonding and complexation (Sharma et al., 2021). One of the key advantages of using biogenic biomass derivatives as adsorbents is their environmentally benign nature, which comes from their ability to reduce waste material without generating toxic byproducts. In addition, biogenic adsorbents are derived from clean and renewable sources, making them both sustainable and cost-effective (Arahman et al., 2019). Biosorption remarkably decontaminates organic, inorganic and persistent pollutants with the help of plant and agricultural wastes (Rusu et al., 2021). Larger ion exchange capability, regeneration/reusability, abundance of adsorption sites accompanied by high surface area, elevated porosity and greater abundance are vital features required to develop an ideal adsorbent (Badawi et al., 2021; Hayat et al., 2023). Plant-based adsorbents are particularly effective in removing anionic dyes, due to the presence of anionic functional groups in their lignocellulosic structure, which create a negative surface charge that enhances adsorption (Haque et al., 2022).

Adsorbents derived from a wide range of organic feedstock, including various biomass sources, municipal waste, agricultural and livestock waste and biochars, provide a cost-effective alternative to activated carbons. The price of biochar-based materials is generally lower, ranging from $350 to $1200 per ton, compared to the price range of $1100 to $1700 per ton for ACs. The cost of biochar-based materials is influenced by multiple factors, including the collection, transport, storage, and pyrolysis of the feedstock, as well as the subsequent storage and transport of these biosorbents (Krasucka et al., 2021). However, it is important to consider the complete lifecycle costs of these biosorbents, their subsequent storage and transport. By optimizing these processes and ensuring efficient logistics, the overall cost of biochar-based adsorbents can be further reduced. Furthermore, the cost-effectiveness of biochar-based adsorbents should be evaluated in conjunction with their adsorption efficiency and overall performance. While biochar-based materials offer a cost advantage, it is crucial to ensure that they maintain high adsorption capacities and exhibit effective pollutant removal.

3. Other strategies for decontamination of drugs and dyes

One of the most effective methods for removing contaminants is the oxidation of pollutant molecules by oxidants, such as H_2O_2. Strong oxidants, such as hydroxyl radical ($OH^{\bullet}$) initiate the advanced oxidation processes. In this method, powerful oxidants cause the breakdown of pollutants into simple, non-toxic substances or complete breakdown to form CO_2 and water. However, the advanced oxidation process is costly due to the low pH adjustment, formation of toxic by-products and the need for quenching the remaining hydrogen peroxide (Deng and Zhao, 2015).

Chemical coagulants, such as lime ($Ca(OH)_2$) and soda ash (Na_2CO_3), are often used to treat wastewater due to their ability to produce rapid and concentrated settle able flocs by chemical coagulation and flocculation. Although this method can increase the efficiency of removing suspended solids compared to plain sedimentation, it has some drawbacks. For example, the use of inorganic toxic coagulants can result in the production of highly toxic sludge and increase operational costs. Additionally, the flocs formed using chemical coagulants can be of poor quality, leading to undesirable results. Furthermore, an increase in pH can reduce the flocculation ability of the flocculent. It is also worth noting that cationic dyes do not coagulate easily (Iwuozor, 2019; Teh et al., 2016).

The removal or reduction of different pollutants in wastewater, such as pharmaceutically active compounds (PhACs), dyes, microorganisms and heavy metals, by filtration membranes, is widely used in industrial and domestic areas. There are different membranes based on pore size, which are classified as microfiltration, ultrafiltration, nanofiltration and reverse osmosis membranes. However, the membrane filtration process has drawbacks, namely, high energy consumption, substantial operational costs, application only to small and medium industries, short lifetime of the membrane and membrane changing before fouling leads to high cost (Obotey Ezugbe and Rathilal, 2020).

Electrochemical treatment is another method used for the removal of dyes and other contaminants from water. This method includes electro-reduction, electro-coagulation and electro-oxidation. These methods are not specific and can be used for several different pollutants. The disadvantages of the electrochemical treatment include the requirement of high initial investment in equipment, maintenance costs and less effective removal of dye due to high flow rates (Roa et al., 2021).

Ion exchange resins are commonly used for the removal of dyes. They are highly selective and easy to regenerate. Ion exchange resins are highly effective for removing organic compounds, including reactive dyes which are typically rapidly adsorbed by these resins. However, ion exchange resins have higher initial and maintenance costs, insufficient effectiveness for dispersing dyes and drugs, generation of secondary pollutants, low durability of resins in the face of high pollution and limited commercial use due to selectivity of resins (Bhandari et al., 2016).

4. Factors affecting adsorption

The rates of biosorption of dyes, pharmaceuticals and other emerging pollutants in aquatic environments are affected by various factors. Research shows that environmental parameters, like temperature, pH, adsorbent dose, agitation rate, initial concentration, contact time, ionic strength and the nature of the adsorbate, can influence adsorption. The characteristics of the adsorbent, including its surface area, particle size and pore structure, also play a critical role in determining the efficiency of the process (Badawi et al., 2021).

The pH of the solution is a critical factor that affects the adsorption of dyes, particularly anionic dyes. Research has shown that the optimal pH for anionic dyes adsorption is typically around pH 2 or 1. This is due to changes in the surface charge of lignocellulose when the pH is lowered. Surface charges of both cationic and anionic compounds are sensitive to pH variations (Haque et al., 2022)). Additionally, the adsorption rate of cationic dyes generally increases at higher pH levels, while the adsorption rate of anionic dyes decreases (Manzoor et al., 2022; Nayagam and Prasanna, 2021).

Temperature is another crucial parameter that impacts the adsorption process. It plays a role in determining whether the process is exothermic or endothermic. High temperatures can enhance the mobility of dye molecules by increasing the biosorption sites of the adsorbent surface or by reducing the solution (Rangabhashiyam et al., 2018).

The contact time between the adsorbent and the solution is also a vital factor that determines the rate of adsorption. However, there is no effect on the adsorption rate after the dye molecules have been deposited on all the available sites.

Moreover, the initial concentration of dye determines the adsorption rate and the initial pollutant concentration has no impact (Badawi et al., 2021; Haque et al., 2022; Nayagam and Prasanna, 2021). The adsorbent dosage also plays a significant role in adsorption, creating excess availability of adsorption sites at a constant dye concentration.

In addition, the pH-dependent interaction between the biosorbent and the pollutant, the net charge on the biosorbent and drug and the degree of ionization of the pollutant are also critical factors to consider (Jureczko and Przystaś, 2021).

5. Plant-based adsorbents for dyes removal

Raw leaves biomass and modified leaves gained significant attention for their potential as adsorbents for the removal of ionic and non-ionic dyes, due to their abundant availability. Leaves contain a diverse array of functional groups, such as amino, carbonyl, nitro, hydroxyl and others, that can interact with the functional groups of dyes, leading to the binding of dyes with leaves (Bulgariu et al., 2019).

The percentage adsorption of dye R (%) was calculated with this equation:

$$R(\%) = \frac{(C_0 - C_e) \times V}{m},$$

The amount of adsorption of dye (q_e) at equilibrium is determined by:

$q_e = \frac{(C_0 - C_e)}{C_0} \times 100$ (Zhou et al., 2022).

Solanum tuberosum peel (STpe) was used to remove bromophenol blue dye from water with the highest adsorption capacity of 8.157 mg/g. The adsorption rate decreased with an increase in pH and dye concentration. Increasing the adsorbent concentration and sonication time caused led to a higher adsorption rate. The hybrid STpe functionalized silver nanoparticles showed a slightly increased uptake of BB from 8.157 to 9.604 mg/g (Akpomie and Conradie, 2020).

Coffee by-products are abundantly available for water purification. The by-products comprise coffee husks, coffee grounds and coffee silverskin. The monosaccharide glucose, galactose, xylose and proteins are the components of coffee residue that offer potential application in the adsorption of dyes (Anastopoulos et al., 2017). Olive stone, generated in olive oil extraction, is a primary by-product.

Renewable biowaste is employed as an essential raw carbonaceous precursor for generating high-quality activated carbon for the elimination of organic pollutants (Saleem et al., 2019). The dead leaves of *Posidonia oceanica (L.)* seasonally accumulate on shore. It is a well known endemic seagrass of vital significance for Mediterranean coastlines. Instead of using expensive commercial activated carbon, their leafs biomass can be used as fixed bed material because of their abundance and high adsorption capacity of 482.6 mg/g for the dynamic adsorption of toxic methylene blue (MB). Moreover, high-quality activated carbon can be prepared from this seagrass species (Cavas et al., 2011). *Posidonia oceanica (L.)* can also be used to adsorb methyl violet (MV) dye (Cengiz and Cavas, 2010).

Aleurites moluccana seeds also show features that allow them to be used as adsorbents. These seeds are effective in dynamic adsorption of hazardous cationic dyes, including rhodamine B (Rh B) and MB, with higher adsorption capacities of almost 117 mg/g and 178 mg/g, respectively (Postai et al., 2016). Also low-cost *Foeniculum vulgare* seeds powder is effective for the adsorption of potassium permanganate ions, due to its high adsorption capacity. Modified *Foeniculum vulgare* seed powder can be used to remove permanganate from polluted water with an adsorption of 89.36% (modified by oxalic acid) (Bani-Atta, 2022).

Pumpkin seed hulls are considered agro-waste, but they are a promising option to produce activated carbon, as they are economical and environmentally effective for efficiently removing erythrosine B, with the highest adsorption efficiency of 16.4 mg/g (Apostol et al., 2016). Loquat is the common name of *Eriobotrya Japonica*; their lignocellulosic seeds can potentially adsorb acid orange in water bodies (Ahmad et al., 2019).

White rice husk ash was used to adsorb brilliant green (BG) dye from an aqueous medium as an ecofriendly, low-cost adsorbent, with a maximum efficiency of 85 mg/g (Tavlieva et al., 2013). Raw *Saccharum munja* (SM) adsorbent and composite functionalized SM can be used for the rapid adsorption of dyes. SM is drought-resistant plant and unsuitable for animal feeding. The presence of cellulose, hemicellulose and lignin provides many functional groups to interact with adsorbates. The reusability of up to 10 consecutive adsorption cycles makes them economical. The synergistic integration of carbon nanotubes (CNT) and SM plants makes them efficient plant-based CNT composite adsorbents for methylene blue and safranin O dyes (Yadav et al., 2021).

Hylocereus undatus, also known as pitaya, is an edible fruit that contains very active compounds. The antioxidant and cytotoxic features of its pulp may be due to the presence of betacyanins, phenolic acids, etc. The peels of pitaya fruit can be used as an efficient adsorbent of alcian blue, MB and other pollutants (Franco et al., 2022).

Lantana shoot and pine needles are considered waste in the environment and difficult to manage; they are used to make effective, low-cost, eco-friendly adsorbents because they have rich carbon content, low ash, low moisture content and the presence of functional groups to interact with an adsorbate (Malik et al., 2015).

BG is a widely used dye that causes acute and chronic effects on human health as well as on animal survival. Various fruit wastes, like banana peel and coconut shell, are potent adsorbents to remove this dye. Kumar and Barakat used binary oxidized cactus fruit peels as an efficient adsorbent for the adsorption of BG (Kumar and Barakat, 2013). Agarwal et al. prepared activated carbon derived from *Peganum harmala L.* seeds to rapidly and effectively adsorb BG from water (Agarwal et al., 2017). This plant is used as a stabilizing agent for ZnO nanoparticles (NPs). The activated carbon generated from *Peganum* can be coated by the prepared ZnO NPs and used for the removal of heavy metals such as chromium (Fazlzadeh et al., 2017).

The adsorptive capacity of powder banana peels was assessed. The presence of carboxyl, amides and hydroxyl groups makes them a powerful adsorbent of few azo dyes, like congo red (CR) and reactive black (both water-soluble anionic dyes) with the highest adsorption rate of 49.2 and 164.6 mg/g, respectively (Munagapati et al., 2018).

The peels of extensively cultivated cucumber, potatoes and bananas were directly used to biosorb anionic and cationic dyes without any modification (Munagapati et al., 2020).

Pistachio is widely produced in Iran, USA and Turkey. Their shells are considered waste but they can be economic biosorbents for basic blue 41 dye, in a continuous and batch operation system (Şentürk and Alzein, 2020).

Other examples of plant-based adsorbent for dyes adsorption can be found in Table S1.

6. Plant-based adsorbents for drugs removal

The active chemical compounds present in pharmaceuticals can persist in water bodies as micropollutants, posing a serious threat to aquatic organisms. These pollutants are found in high concentrations in wastewater, groundwater and even drinking water, particularly in the case of commonly used nonsteroidal anti-inflammatory drugs, like ibuprofen and paracetamol. The presence of these substances in water can lead to serious health issues, such as gastrointestinal and kidney problems, in humans.

To address this problem, researchers have explored various methods for removing pharmaceuticals from water sources. One promising approach is phyto remediation, which uses plant extracts to remove emerging drugs from wastewater (Gałwa-Widera, 2019). Another approach involves the use of porous activated carbon made from plant bark as an adsorbent to remove pharmaceutical drugs like naproxen from watercourses (Franco et al., 2022).

The COVID-19 pandemic led to a significant increase in the generation of biomedical waste, particularly from the use of analgesics for pain relief. This contributed to the already pressing issue of pharmaceutical pollution in water bodies. Triclosan, a common ingredient in antibacterial soaps and other personal care products, is an emerging pollutant that poses a global threat to the environment. It is found in high concentrations in water bodies and can cause significant harm to aquatic life.

To solve this problem, researchers have been investigating the use of natural biosorbents to remove pollutants from water sources. One promising option is *Sargassum*, a type of alga that effectively adsorbs dyes and drugs from aquifers. This method is highly efficient and cost-effective, making it a practical solution for water treatment (López-Miranda et al., 2022).

Another natural biosorbent is the *Moringa oleifera Lam.* plant, commonly used as a low-cost option for water treatment. The seed husks of this plant, combined with iron oxide nanoparticles, have good adsorptive capacity for triclosan and other pollutants (Cusioli et al., 2021).

The presence of fluoroquinolone antibiotics, such as norfloxacin and ciprofloxacin (CIP), in aquatic systems is a growing concern, due to their potential harm to aquatic life and human health. To address this issue, researchers investigated the use of Long-root *Eichhornia crassipes* activated carbon as an adsorbent to remove these antibiotics from water sources (Liu et al., 2019). This approach showed promising results in effectively reducing the presence of these pollutants in aquatic systems.

Another promising method for removing pharmaceuticals from water is the use of biochar. In particular, pinewood biochar nanoparticles are highly effective for adsorbing the carbamazepine pharmaceutically active compound from water. This approach has the potential to remove this harmful substance from wastewater, with nano biochar adsorbing up to 95% of carbamazepine (Naghdi et al., 2019).

Walnut shells showed great promise as adsorbents for the widely used antibiotic cephalexin, with a maximum adsorption efficiency of 233 mg/g (Jahanban-Esfahlan et al., 2020). This natural material could provide a low-cost and environmentally friendly solution for the removal of this pollutant from water systems.

Heterotrophic bacteria were also used to modify wheat straw and create an effective adsorbent for perchlorates, achieving an impressive adsorption efficiency of 119 mg/g (Tan et al., 2012). Another innovative approach is the use of the *Scripus validus* plant, which is able to remove up to 98% of naproxen and almost 74% of carbamazepine at a concentration of 0.5–2 mg/l (Zhang et al., 2013).

Jacaranda mimosifolia seed pods were also investigated as a source of activated carbon for removing the nonsteroidal anti-inflammatory drug ketoprofen from water systems, achieving a maximum removal efficiency of 96% due to their high pore volume and surface area (Georgin et al., 2021).

The seeds of avocado were evaluated for their potential as an activated carbon source, with promising results for the removal of multiple hazardous phenolic and pharmaceutical products. Optimizing the pyrolysis time and temperature can increase the functional groups and surface area of the resulting activated carbon (Leite et al., 2018).

Nauclea diderrichi biomass activated carbon, derived from naturally grown sources, is promising to remove pollutants from water samples. H-bonding, π-π and electrostatic interactions were the observed mechanisms behind the removal of ibuprofen and MB, with maximum removal efficiency of 70.92 mg/g and 35.09 mg/g, respectively (Omorogie et al., 2021).

Peanut shells modified with potassium ferrate showed potential for synthesizing partially graphitic biochar, which is effective in removing diclofenac and ibuprofen from water samples (Nguyen et al., 2021).

Tomato waste was investigated for its ability to remove tylosin, dexamethasone and praziquantel (Mutavdžić Pavlović et al., 2021). The study revealed that the adsorption was spontaneous and endothermic due to the negative enthalpy change value.

Litchi peels bio waste were used to fabricate magnetic porous iron-based materials to remove pharmaceutical amaranth dye, with a maximum adsorptive efficiency of 44.87 mg/g through pyrolysis (Foletto et al., 2017).

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Several eco-friendly approaches using natural sources are also promising for the removal of pollutants from water samples. Some examples are shown in Table S2.

7. Physical and chemical modification of biogenic adsorbents

Biogenic adsorbents can be physically, chemically and thermally modified to enhance their adsorptive capability for dyes and drugs (Haque et al., 2022; Sharma et al., 2021). Physical pretreatments such as autoclaving, heating, boiling, grinding and carbonization can influence the functional groups on the adsorbent surface, particle size and surface area (Ahmad et al., 2019).

On the other hand, chemical modification can improve the ability of

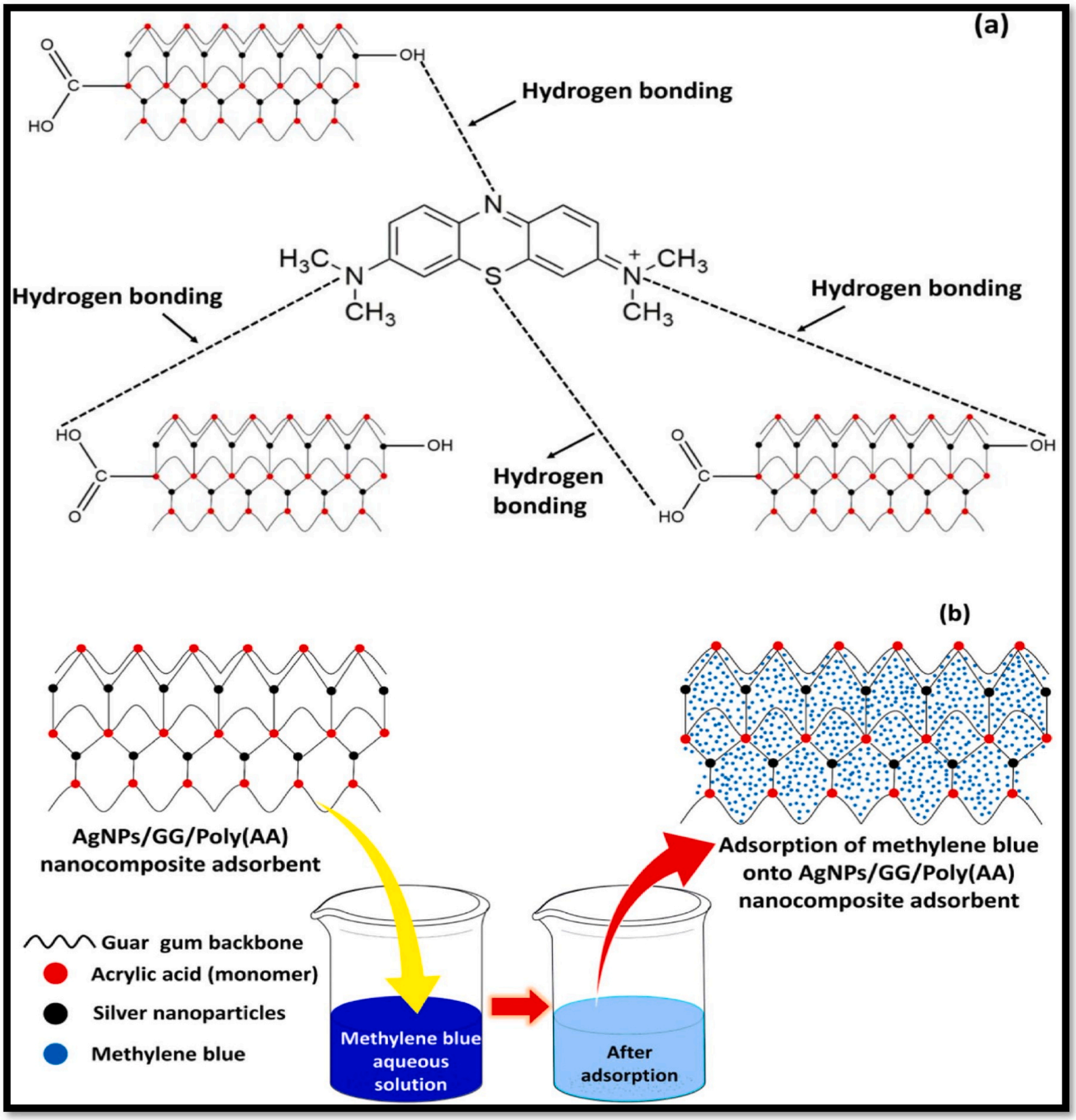


Fig. 2. Mechanism of AgNP/AA/GG biosorbent for the uptake of MB. Reprinted from (Singh and Dhaliwal, 2021) with permission from Springer.

the adsorbent to extract soluble organic compounds. Modifying agents like base and acid solutions, oxidizing solutions and organic compounds are employed to decolorize and degrade dyes, drugs and heavy metals (Ngah and Hanafiah, 2008). Acid treatment increases microporosity and surface area of the adsorbent (Eletta et al., 2020), while functional groups, particle surface, and particle affinity play a significant role in trapping contaminants within interstitials on a macromolecular matrix (Reddy et al., 2011). Chemically modified biosorbents possess an improved specific surface area, which can range from 700 to 1400 m^2/g for commercially available activated carbon (Abdullah et al., 2001). Additionally, nanomodification is a promising method to enhance the performance of biosorbents (Khadir et al., 2020). Other authors reported Ag nanoparticles containing grafted synthetic monomer acrylic acid on guar gum biopolymer nanocomposite for the effective removal of MB (mechanism representation in Fig. 2) (Singh and Dhaliwal, 2021) (see Fig. 3).

For example, the core of the *Artocarpus camansi* fruit, that has no economic value, can be chemically modified with a dilute sodium hydroxide solution to significantly enhance its ability to adsorb dyes. Breadnut skin, modified in this way, increased its adsorption capacity for MG to 353 mg/g and its reusability to more than five cycles (Chieng et al., 2015). Similarly, strong NaOH-modified *Artocarpus odoratissimus* leaves demonstrated increased adsorption activity for MG, rising from 245.9 to 422.0 mg/g after modification. (Zaidi et al., 2019).

Cellulosic sisal, also known as *Agave fourcroydes* and *Agave cantala*, is another plant material used to remove heavy metals and various dyes from water. Modification of the cellulosic sisal fibre can lead to lower adsorbent dosage and more efficient performance. A recent study utilized in situ chemical oxidative polymerization to modify cellulosic sisal fibre with polypyrrole-polyaniline nanoparticles, resulting in a composite material with an impressive adsorptive capacity of 88% for ibuprofen (Khadir et al., 2020).

Date pits, a common agricultural waste material, were also used as a biosorbents for the removal of dyes from water. $ZnCl_2$-activated date pits were more effective than raw date pits, with an increase in adsorption activity for both MB and methyl orange (MO) from 45 to 27 mg/g to 403 and 301 mg/g, respectively (Mahmoudi et al., 2015).

In a study, researchers conducted magnetic modification of *Leptothrix* sp. Sheaths using nano and micro particles of iron oxides and ferrofluid (Angelova et al., 2017). This magnetic modification proved to be successful in enhancing the uptake of amido black 10 B at a pH of 2, harnessing the magnetic properties of the modified sheaths. Furthermore, the efficiency of various biosorbents for the removal of the MB dye was compared, with magnetic *Cortaderia selloana* flower spikes (nM∞CSFs) demonstrating an excellent biosorption capacity of 119.05 mg/g, surpassing the adsorption capacity of 72.99 mg/g exhibited by the unmodified *Cortaderia selloana* flower spikes (CSFs) (Parlayıcı and Pehlivan, 2021). Additionally, alginate-immobilized *Saccharomyces pastorians* and *Saccharomyces cerevisiae* yeast strains were synthesized as effective biosorbents for the adsorption of organic pollutants. The immobilized biosorbents exhibited efficacy in eliminating dyes such as indigo carmine and orange II, as well as drugs including ethacridine lactate, cephalexin, and rifampicin. The maximum adsorption capacity depended on the specific type of natural polymer used for immobilization (Rusu et al., 2021).

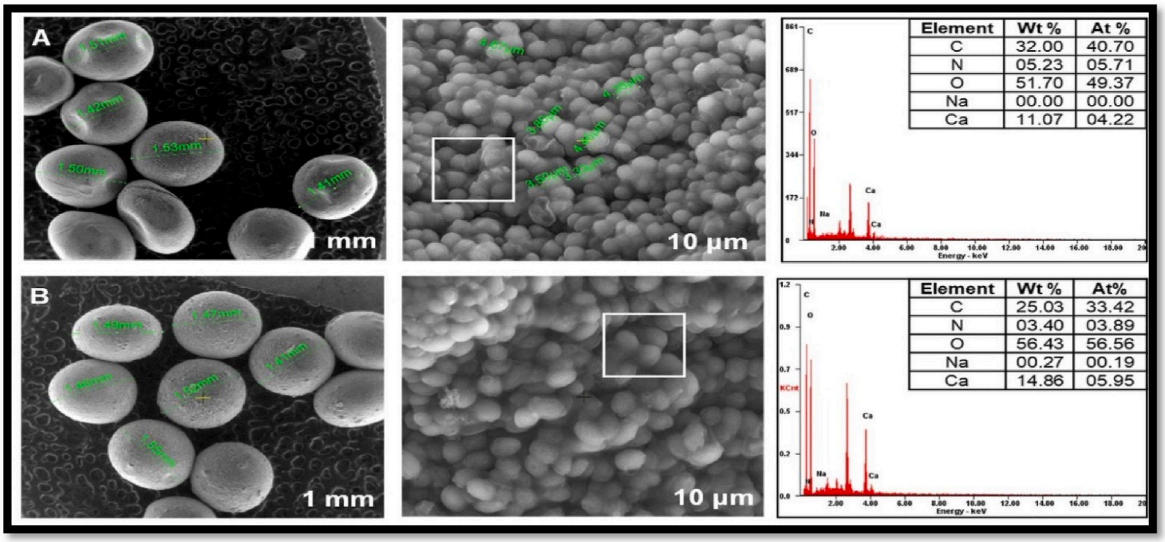
Albizia lebbeck seed pods modified with KOH showed increased adsorption efficiency for the removal of cephalexin, with an adsorption capacity of 137.02 mg/g compared to 118.08 mg/g for K_2CO_3 modified seeds (Ahmed and Theydan, 2012).

Overall, these studies demonstrate the potential for plant-derived biosorbents to be effective and low-cost options for removing pollutants from water, and the importance of chemical and physical modification in optimizing their performance. Some examples are given in Table S3. Fig. 4 shows a scheme of activated carbons derived from plants, activation methods adsorption processes and mechanisms.

8. Plant-based nano adsorbents

Environmental remediation seems promising with the use of nano adsorbents, due to their unique properties, such as high porosity, large surface area, small particle size, catalytic potential and abundant reactive sites. The synthesis of nanoparticles is a rapidly growing field of research that allows for controlled particle morphology and crystalline structure, resulting in enhanced adsorption efficiency of plant-based adsorbents (Sharma et al., 2019). Carbon-based nanostructured materials are effective for water purification, but they come with a high cost and potential toxicity concerns, making plant-based nano adsorbents more environmentally friendly with efficient degradation activity (Aguilar-Pérez et al., 2020). Cellulose, the primary ingredient in plants, has natural adsorptive properties, due to its multiple functional groups. By reducing its size to the nano range, its adsorption capability can be further improved by increasing the surface area (Aguilar-Pérez et al., 2020; Sahoo and Prelot, 2020).

Integrating biogenic materials with advanced materials, such as



Element	Wt %	At %
C	32.00	40.70
N	05.23	05.71
O	51.70	49.37
Na	00.00	00.00
Ca	11.07	04.22

Element	Wt %	At%
C	25.03	33.42
N	03.40	03.89
O	56.43	56.56
Na	00.27	00.19
Ca	14.86	05.95

Fig. 3. SEM images of alginate immobilized (a) *Saccharomyces cerevisiae* and (B) *Saccharomyces pastorians* yeast strains. Reprinted from (Rusu et al., 2021) (open access Creative Commons CC BY 4.0 license from MDPI).

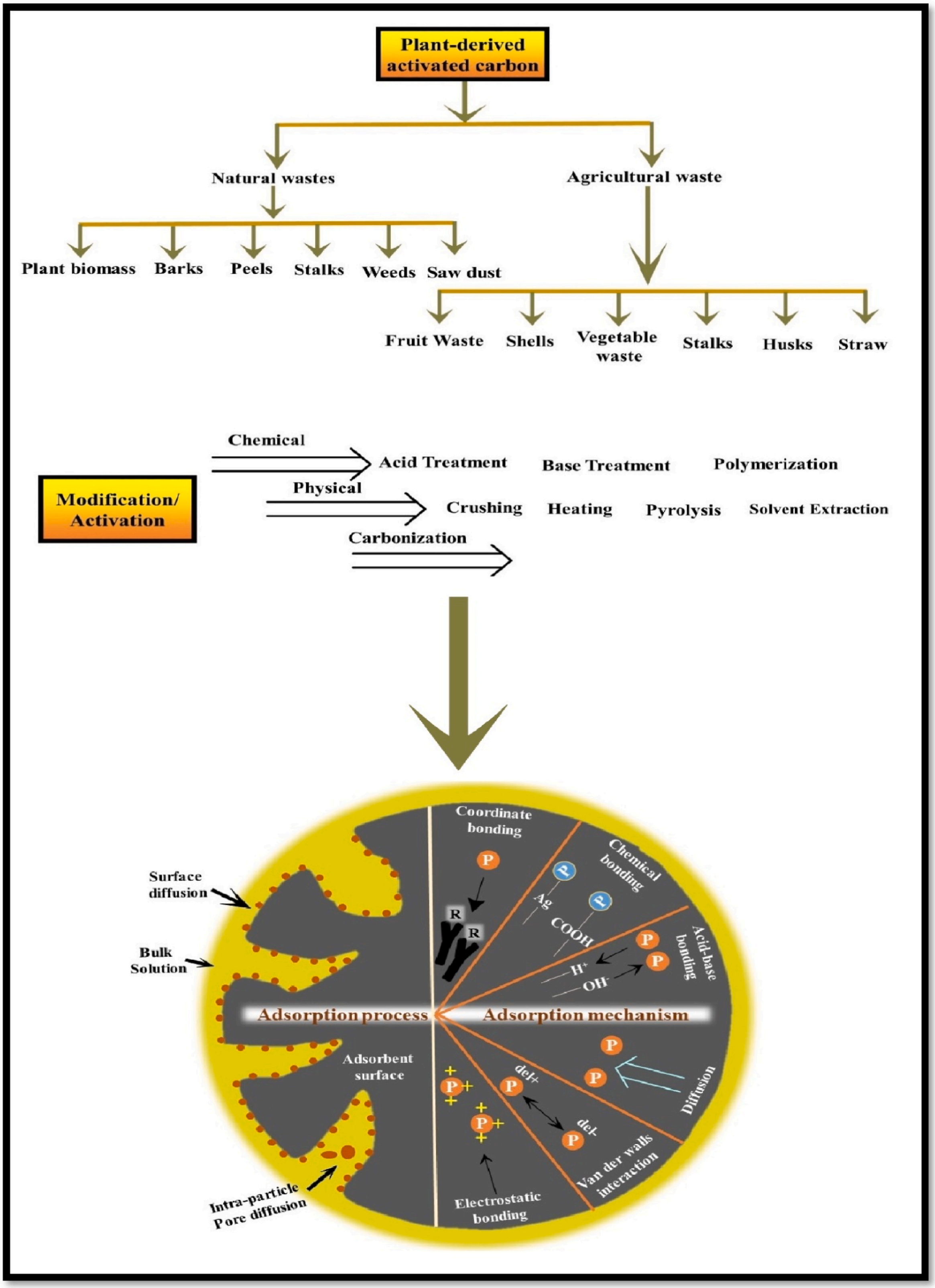


Fig. 4. Schematic representation of plants derived activated carbon, their activation method adsorption process and mechanism (redrawn and adapted from (Badawi et al., 2021; Goswami et al., 2022)).

nanoparticles, offers interesting potential applications, due to their combined mechanical stability, specificity and large surface area. The coupling of plant-based materials with nanomaterials provides unique properties that exceed those of traditional materials as adsorbents (Shaikh et al., 2021). Comparison of plant (raw lignocellulose) adsorbent with lignocellulosic based composite and the production of these composite are given in Fig. 5.

Several nanostructured materials were studied as potent candidates for removing dyes and drugs from wastewater (Nayagam and Prasanna, 2021). Nanomagnetic particles with high surface area and extended reactivity show great efficiency as reducing agents, to remove various dyes from water bodies. However, the easy aggregation and precipitation of nanoparticles limit their applications. Recent studies demonstrated that plant-based materials prevent agglomeration and extend the reactivity of nanoparticles (Parlayıcı and Pehlivan, 2021).

Plant-based materials emerged as sustainable and efficient ways to synthesize nanoparticles, due to their large amounts of antioxidants that can reduce metal ions. For example, the bud extract of *Syzygium aromaticum* was used to synthesize iron oxide nanoparticles for the efficient removal of MB dye (Jain et al., 2021), while Fe nanoparticles synthesized from tea extract were employed to decolorize MG dye with a maximum activity of 90% (Gautam et al., 2019).

In order to deal with the adsorption of anti-inflammatory diclofenac and antibiotic CIP, researchers scaled down cellulose obtained from *Cyperus rotundus* grass to nanocellulose, achieving a maximum adsorption of 192.30 and 227.22 mg/g, respectively (Shahnaz et al., 2021). *Mentha pipertia* L. plant was a proficient and environmentally benign source for synthesizing silver nanoparticles as nano adsorbents for the low-cost, scalable method to remove CV dye with a high adsorption efficiency of 704.07 mg/g (Moghadas et al., 2020).

Additionally, mango seeds were studied for their adsorption characteristics of the anti-cancer drug doxorubicin hydrochloride at different contributing parameters (Altalhi et al., 2022).

The integration of plant-based materials with advanced materials, such as nanoparticles, presents a potential solution for enhancing the stability and reactivity of adsorbents in wastewater treatment. This promising approach aligns with the growing demand for environmentally-friendly and effective treatment methods. Table S4 shows some examples.

9. Plant based composites as adsorbents

Fig. 6 shows different biogenic sources used in the biosynthesis of composites and nanoparticles. Several studies explored the use of various plant composites for the removal of dyes and other contaminants. A novel green metal-organic framework (MOF) nanocomposite based on cucumber peel activated carbon (AC) and chromium based MOF (MIL-101(Cr)) was studied for the elimination of reactive yellow 186 and acid green 25 dyes (Mahmoodi et al., 2019). Titanium oxide nanoparticles on watermelon rind-derived AC were evaluated for the ultrasonic-assisted degradation of water-soluble CR and phenol red anionic dyes (Masoudian et al., 2019). Tamarind-based nanocomposites are also efficient for the removal of several dyes under various conditions, as recently reviewed (Malik et al., 2022).

Nigella sativa, also known as black cumin (BC), contains a variety of functional groups, which make the seed surface highly suitable for the adsorption of charged ions. $MnFe_2O_4$/BC is a hybrid composite obtained by co-precipitation, with antibacterial activity against both Gram-positive and Gram-negative bacteria, able to adsorb MB (Siddiqui and Chaudhry, 2018). A magnetic nanocomposite of iron oxide nanoparticles on quince seed mucilage, synthesized by in situ formation, also showed efficient performance for the adsorption of MB (Hosseinzadeh and Mohammadi, 2015).

The aqueous seed extract of *Carum carvi* L. seeds can be used as a capping, stabilizing and reducing agent to reduce aggregation and precipitation during Fe_2O_4/Au nanocomposite synthesis for the photocatalytic degradation of antibacterial drug imipenem and anticancer drug imatinib (Mirsadeghi et al., 2020). *Shorea robusta* (Sal tree) leaves were used to synthesize silver nanoparticles and biochar. Both were coupled through coprecipitation to form a porous, stable and heterogeneous biochar-based silver nanocomposite for the biodegradation of CR and Rh B dye with an adsorption efficiency of about 80% (Shaikh et al., 2021).

Lemonwood was used to derive activated carbon which is used to adsorb dyes. The coupling of lemonwood AC (ACL) and iron oxide nanoparticles to form nanocomposite was more effective for CV removal

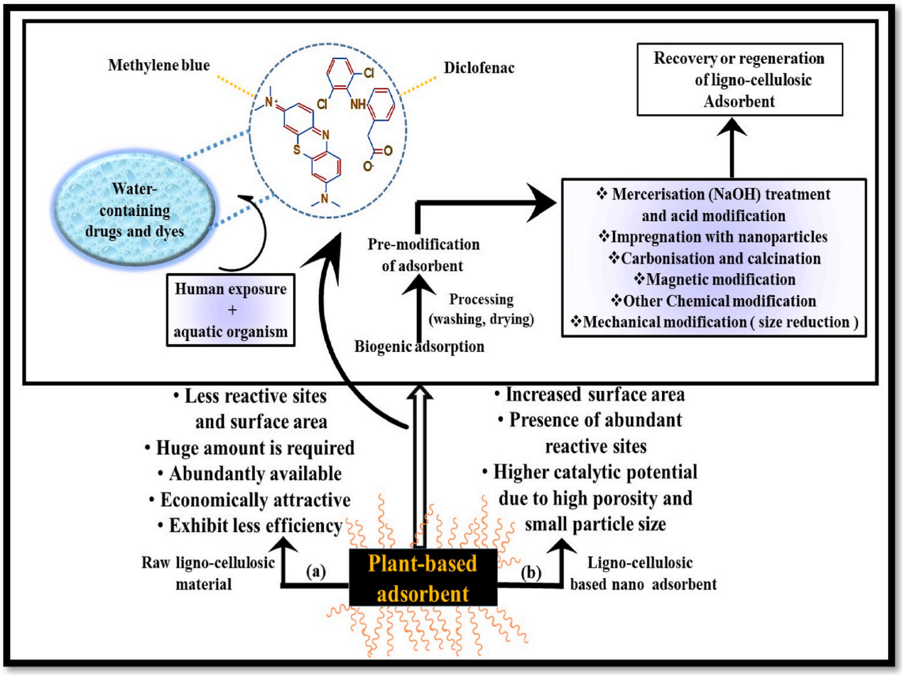


Fig. 5. Environmentally benign and economical production process of plant (lignocellulose) based adsorbent.

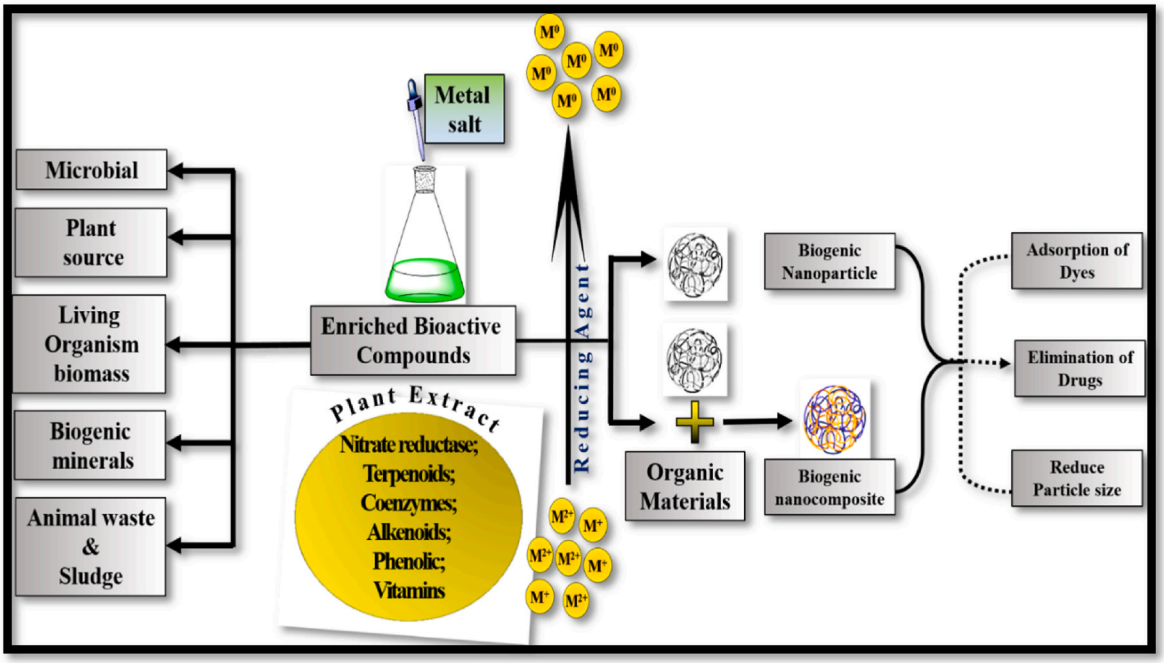


Fig. 6. Different biogenic sources used in the biosynthesis of composite and nanoparticles.

from an aqueous solution at pH 9. The highest adsorption capacity of CV increased from 93.5% (ACL) to 98.3% (ACL/Fe_2O_3) (Foroutan et al., 2021).

The fusion of sugarcane ash with thermoplastic polyethylene terephthalate polymer was evaluated for the saturation of naproxen and diclofenac. The maximum saturation of diclofenac and naproxen on active sites of hybrid composite reached 324.34 μg/g and 956.49 μg/g with a saturation time of 22 and 8 h, respectively (Américo-Pinheiro et al., 2022).

10. Biogenic minerals

Hydroxyapatite (HAP) is a biogenic mineral that can be produced from several sources (Fig. 8). Bhushan et al. combined HAP with chitosan (CT) and magnetite to form a hybrid organic-inorganic composite that enhanced the adsorption of CIP from wastewater (Bhushan et al., 2022). The composite adsorbent exhibited the highest adsorption efficiency of 516.98 μg/g, which is superior to the values for the individual components.

In a study conducted by Thakur et al. the adsorption capacity of calcium oxide nanoparticles derived from eggshells was compared to that of commercial calcium oxide for the removal of phenol red and brilliant green dyes (Thakur et al., 2022). The researchers also investigated the adsorption behavior in a binary mixture of both dyes, as depicted in Fig. 7. The findings revealed that the eggshell-extracted calcium oxide exhibited superior surface characteristics and demonstrated promising adsorption performance compared to the commercial counterpart. In the binary mixture, the eggshell-derived calcium oxide showed an impressive uptake of 78% for phenol red and 98% for brilliant green dye.

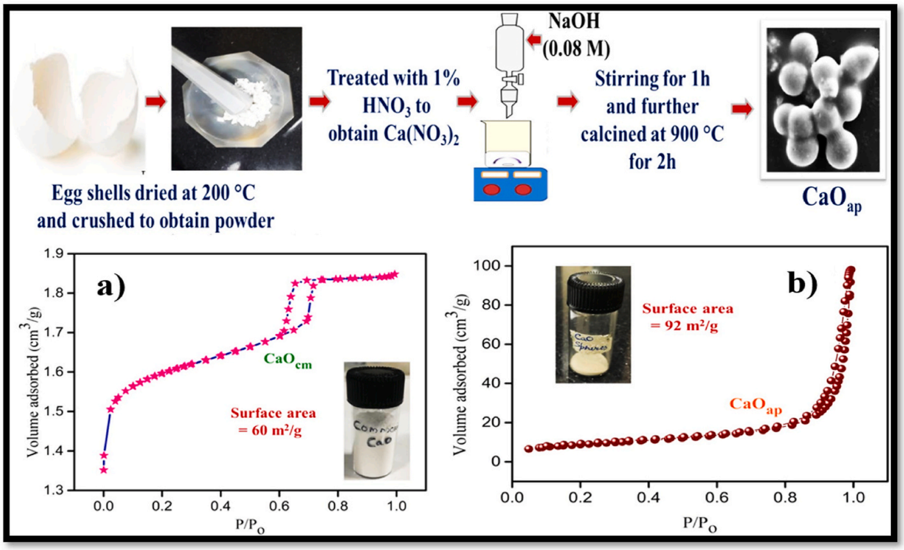


Fig. 7. Schematic synthesis of egg shell derived CaO and comparison of its surface area with commercial CaO. Reprinted and adapted from (Thakur et al., 2022) with permission from Springer.

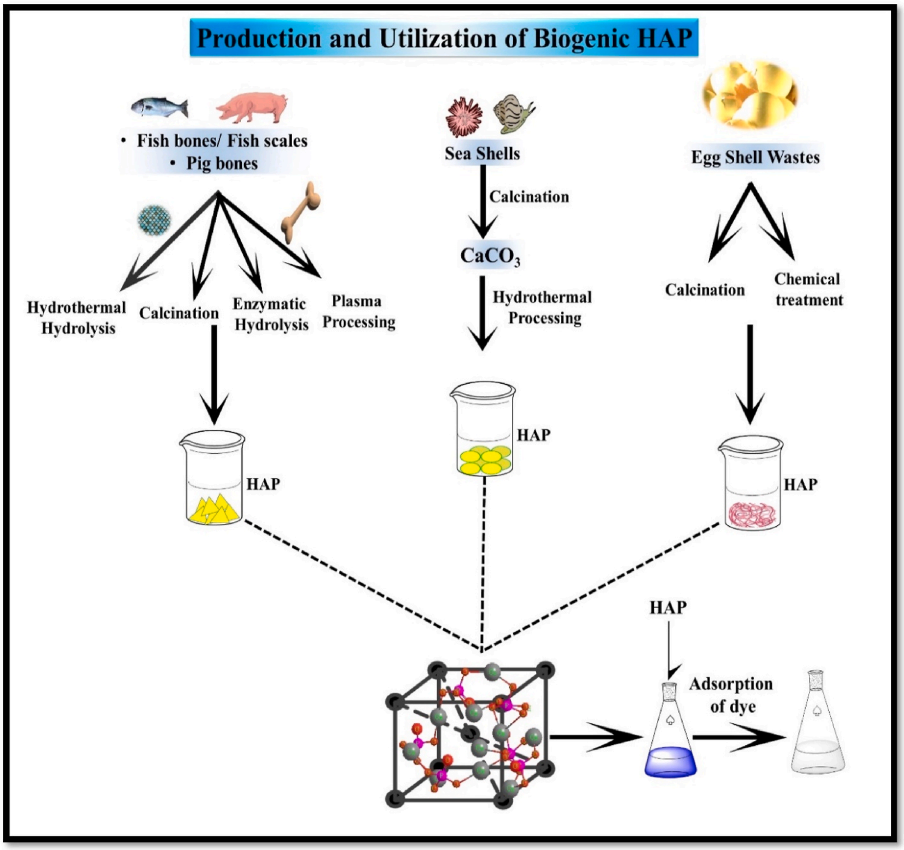


Fig. 8. Major production methods and uses of HAP (adapted from (Aiza Jaafar and Zainol, 2023)).

In another study, a ternary nanocomposite of magnetic functionalized HAP and MIL-101(Fe) MOF was prepared by a hydrothermal approach to evaluate the adsorption of antibiotics tetracycline (TC) and CIP from water samples. The maximum removal efficiency for TC and CIP on HAP/MIL-101(Fe)/Fe_3O_4 was 95% and 93%, respectively, in neutral pH conditions, with the adsorption process being a spontaneous endothermic reaction (Beiranvand et al., 2022). A silver nanoparticle functionalized HAP adsorbent was effective for MB and CR dye adsorption, with adsorption parameters, such as pH, dye concentration, temperature and adsorbent concentration significantly influencing its adsorption characteristics (Azeez et al., 2022). Natural zeolites, including heulandite, phillipsite and clinoptilolite, were applied for the adsorption of safranin dye pollutants over a wide range of parameters. The adsorption capacity of these selected zeolites for cationic dyes is higher than that for anionic dyes (Abukhadra and Mohamed, 2019). A greenway synthesized magnetic perlite iron oxide nanocomposite was modified with organic ibuprofen to enhance its adsorption efficiency for direct Red-81, increasing it from 142.85 to 416.66 mg/l at pH 4–6 (Shirkhodaie et al., 2016). Cr(VI)-imprinted poly (4-VP-*co*-MMA) (IIP) supported on activated Indonesia natural zeolite was observed to selectively adsorb Cr(VI) in the presence of other competetive metal ions such as Cr (III), Mn(II), Pb(II) and Ni(II) (Neolaka et al., 2018).

11. Algae and microorganisms for dyes and drugs removal

Table S5 presents several microorganisms and algae that can effectively remove drugs and dyes from polluted water. Algal biomass contains cellulosic polysaccharides, proteins and lipids, making it a promising biosorbent for various harmful pollutants. The presence of carrageenan and alginate in algal polysaccharides enables them to exhibit exceptional binding affinity to different pollutants. Microbial gum, derived from fermentation of specific microorganisms, and marine gum, present in various algal cell walls, are also effective adsorbents of pollutants (Sharma et al., 2018).

In a recent study, alkaline-modified *Scendesmus obliquus* biomass was used to remove tramadol from wastewater, by hydrophilic interaction of hydroxyl and carbonyl groups present on the surface of modified algal biomass with the carbonyl and amino groups of the active pharmaceutical compounds (Ali et al., 2018; Mokhtar et al., 2017). In another study, the marine algae *Euchema spinosum* proved to be a sustainable candidate for the adsorption of MB dye. Under ambient conditions, the active sites on the algae were entirely saturated with the dye, indicating an excellent affinity of 0.017 towards MB binding (Mokhtar et al., 2017).

Spirulina algae nanoparticles were developed as an eco-friendly adsorbents for direct yellow 12 (DY12) in water. The adsorption isotherm study showed that spirulina could adsorb up to 714 mg/g of DY12 at acidic pH (Marzbali et al., 2017).

In another study, two types of marine algae, *Macrocystis integrifolia bory (S12)* and *Lessonia nigrescens bory (L13)*, were evaluated for their potential as eco-friendly water treatment options for sulfamethoxazole (SMX) and sulfacetamide (SAM) drugs. The adsorption mechanism was driven by hydrogen bonding at pH 6 and 7 for L13 and S12, respectively. The adsorption isotherm study indicated that L13 was more effective in removing SMX, while S12 was more efficient in removing SAM (Navarro et al., 2014).

Activated carbon derived from filamentous algae was magnetized with Fe_3O_4 to form a magnetic nanocomposite, which was used to remove cephalexin from water. The optimum parameters for this process were determined to enhance its efficiency (Afshin et al., 2020).

Living microalgae *Chlorella* sp. cells were also found to be effective biosorbents for diclofenac and ketoprofen. Different isotherm models were used to describe the adsorption mechanism, with hydrogen bonding and π-π interactions being the most proeminent ones. At pH 6, the maximum adsorption capacities were 0.429 and 0.328 mg/g, respectively (Hifney et al., 2021). Another study compared the adsorption capacity of living and dead biomass of freshwater microalgae *Chlorella vulgaris* against the anticancer drug flutamide. Under optimized conditions, the living cells proved to be a better biosorbent than the dead biomass (Habibzadeh et al., 2018).

A highly efficient amine crosslinked starch (ACS) adsorbent was developed for the selective removal of anionic organic contaminants, including amaranth, diclofenac sodium and brilliant blue, due to the porous microstructure and high density of the amine groups of ACS (shown in Fig. 9). The isotherm studies showed a maximum removal capacity of 724.6, 595.2, and 1287.7 mg/g, respectively (Gao et al., 2021) (see Fig. 10).

Bacterial cellulose has a porous structure, high tensile strength, excellent loading, elasticity and higher removal activity, with rapid biosorption ability (Zhang et al., 2019).

Naturally available biomass from *Phellinus adamantinus* and *Daedalea africana* fungi was studied for the adsorption of MB, with the maximum adsorption capacity being 1.8387 and 0.5210 mol/kg, respectively (Sintakindi and Ankamwar, 2020). *Phanerochaete chrysosporium*, a white rot fungus, was used as precursor for the synthesis of magnetic biocarbon by cultivation in a Fe medium. The obtained iron-rich biomass exhibited high adsorption efficiency of 361.25 mg/g for diclofenac (Luo et al., 2019).

Sewage sludge, containing multiple drug-resistant bacteria such as *Enterococcus*, *Staphylococcus* and *Enterobacteriaceae*, was evaluated for the removal of diclofenac. The activated sludge exhibited over 80% adsorption for an initial concentration of 1 mg/l of diclofenac (Elshikh et al., 2022).

A polyethylenimine-caged platinum nanocomposite was successfully synthesized on bacterial cellulose (PEI-PT@BC) by a facile in situ reduction approach. This led to a versatile fabrication of an ideal biosorbent for MB and anionic acid black dye. While the maximum MB adsorption capacity was only 13.5 mg/g, PEI-PT@BC showed the highest performance of 1157.9 mg/g for anionic acid black dye (Huang et al., 2020).

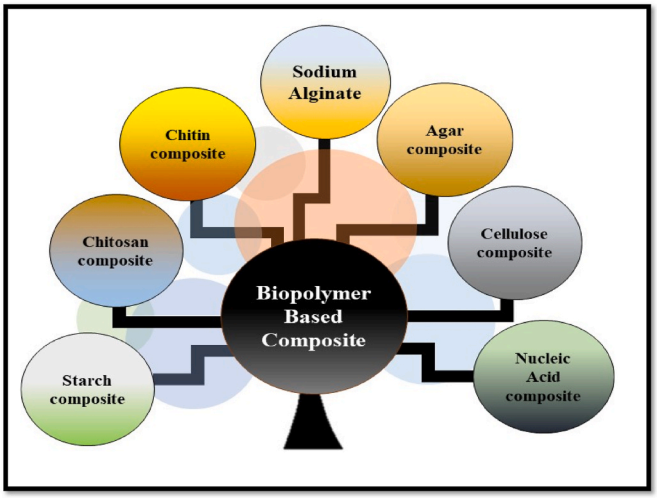


Fig. 10. Multiple biogenic polymers used as adsorbent of drugs and dyes.

A novel hybrid nanocomposite was developed by including sodium zeolite on bacterial substrate (*Gluconobacter oxydans*) for the uptake of CR and MB from an aqueous medium (Ibrahim et al., 2022). A biohybrid composite of natural silk protein sericin and HAP was able to efficiently remove CR dye from water samples. The morphological integrity of the composite allowed for the easy separation of the biohybrid adsorbent from water containing free-flowing pollutants (Koley et al., 2016).

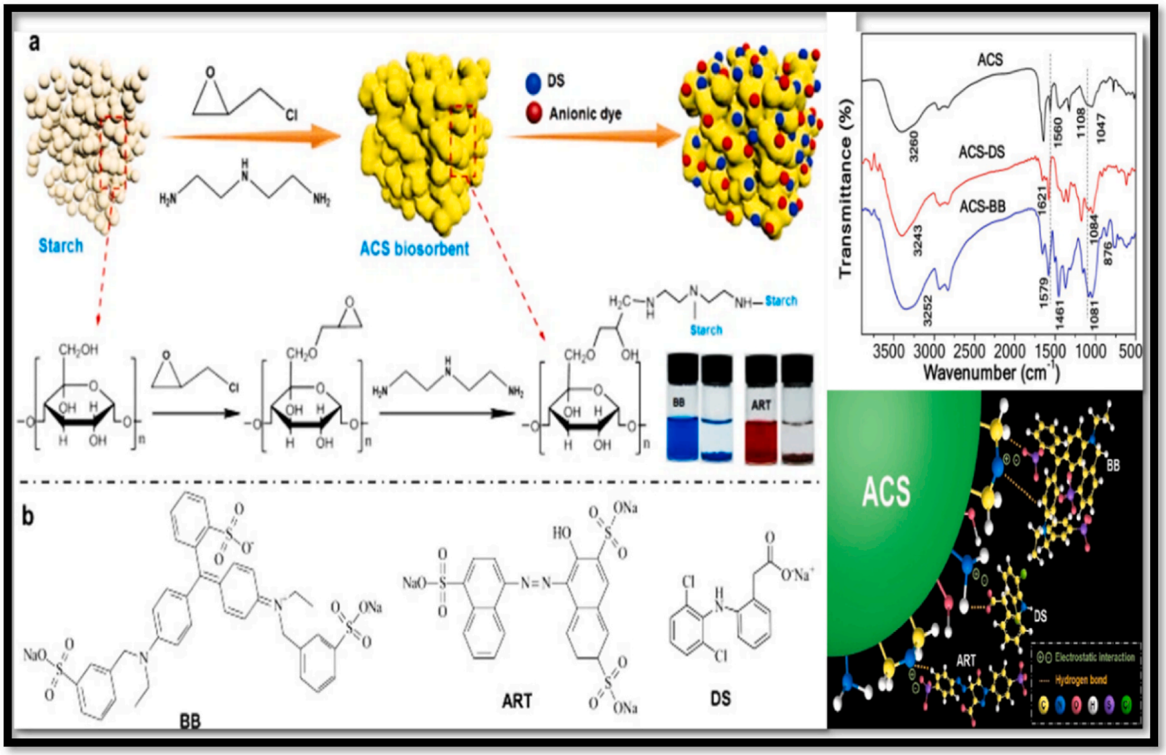


Fig. 9. Schematic representation of amine crosslinked corn starch for adsorption of diclofenac sodium, brilliant blue and amaranth with their molecular structures, FTIR spectra of sorbent before and after removal of DS and BB and their adsorption mechanism. Preprinted and adapted from (Gao et al., 2021) with permission from Elsevier. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

12. Biopolymers for the adsorption of dyes and drugs

Biopolymers are extensively used as adsorbents for the removal of pharmaceuticals and dyes (Fig. 6 and Table S6) due to their availability, biodegradability and low cost. Polysaccharides, natural polymers, terpenes and lipids are reported for their ability to adsorb organic and inorganic contaminants (Kaur et al., 2020). The presence of hydroxyl, ketones and carboxylic groups in biopolymers enables them to interact with the pollutant molecules (Kumar et al., 2022). Chitin, the second most naturally occurring polymer after cellulose, is widely used for the adsorption of different classes of drugs and dyes, due to its unique structure, enriched with hydroxyl and amino groups (Ahmed et al., 2020). Chitin can be extracted from various sources, like microorganisms (brown and green algae, spores), insects (spiders, scorpions, beetles) and the crust of sea animals.

Various synthetic and natural materials, such as biomass, clay, resin, silica and polymers, can be incorporated to modify the pore characteristics and functionality of biopolymers (Ahmed et al., 2020). Functionalization of biomolecules is an effective way to modify their adsorption capacities. Recent studies explored the use of various biopolymers and their derivatives for a wide range of applications, including as nanocomposites, support matrix, catalyst and surface capping agents (Jiang et al., 2009; Preethi et al., 2017). Alginates, for example, are linear block copolymers that are commonly used in wastewater treatment due to their enriched surface functionalities, high water permeability and ability to form sustainable hydrogels (Nasrollahzadeh et al., 2021). Cellulose and alginate are also commonly used as precursors for the synthesis of biogenic composite adsorbents.

Thermodynamic studies showed that synthetic polyamidoamine impregnated on CT backbone is highly effective for the biosorption of anionic amido black 10 B and CR dye (Banisheykholeslami et al., 2021). In addition, naturally derived gums that can be considered as polysaccharides, including seed gum, microbial gum, marine gum and plant exudate gum, can also be used as supports, binders and adsorbents (Sharma et al., 2018).

In addition to biopolymers, protein/PVA nanofibers extracted from *Morinaga stenopetala* seeds were developed for the effective adsorptive removal of carbamazepine and acidic NSAIDs. The maximum adsorption capacity of 333.33 mg/g was obtained, and the interaction between the nanofibers and pharmaceuticals was predominantly ruled by chemisorption (Kebede et al., 2019).

Lignin-based adsorbents also gained considerable attention due to their affinity with certain pollutants (Wang et al., 2022). For instance, a sodium alginate-grafted poly acrylic acid nanocomposite was prepared by a free radical graft copolymerization approach, offering excellent adsorption of MV dye from an aqueous solution (Thakur and Arotiba, 2018).

Polysaccharide hydrogels are able to adsorb massive quantities of contaminants, attributed to their diversity of functional groups, such as epoxy, carbonyl, hydroxyl and amino groups. The polymeric substrate gives permeability, physicochemical residence and multifunctionality because of its 3-D chemical or physical cross-linking (Zhao et al., 2023).

Salecan, a non-ionic microbial polysaccharide comprised of a linear chain of D-glucose units linked by glycosidic linkage, was used to design a hydrogel for the uptake of MV from water. This hydrogel offers controlled pore size and high elasticity, leading to the highest adsorption efficiency of 178.9 mg/g (Qi et al., 2019).

A magnetic MOF encapsulated starch hydrogel (NFe_3O_4@Zn (GA)/starch hydrogel) was assembled for the adsorptive removal of fluvastatin drug from a water sample. The maximum adsorption capacity of 782.05 mg/g was obtained at pH 10 (Mohamed and Mahmoud, 2020a).

Hen feathers, which have high crude protein content, were effective to remove CR dye. The process was endothermic, spontaneous and highly dependent on factors such as pH, contact time and concentration (Mittal et al., 2014).

In another study, a high-value cellulose-derived carbon nanosphere was synthesized by a one-pot microwave-assisted carbonization approach, which showed promising adsorption of commonly detected drugs (diphenylamine, benzophenone, diclofenac, and ketoprofen) due to prominent hydrogen bonding and π-π stacking interactions (Feng et al., 2018)

A reproducible and novel Fe_3O_4/activated charcoal/cyclodextrin/sodium alginate composite was assessed for the saturated adsorption of MV (5.88 mg/g) > ciprofloxacin (3.125 mg/g) > norfloxacin (2.551 mg/g) > BG (5.88 mg/g) (Yadav et al., 2021).

Amine-functionalized carbon microspheres, derived from glucose, were evaluated for their adsorptive capacity towards anionic dyes tartrazine and methyl orange through electrostatic interaction. The kinetic studies were fitted with Langmuir isotherm (Liu et al., 2022). Frozen alginate-montmorillonite beads were prepared from natural polysaccharides and clay, which were effective in physically adsorbing MB from water (Uyar et al., 2016).

Gelatin, a denatured form of collagen, was used as an adsorbent due to its carboxyl, amino and hydroxyl group-enriched structure, that allows for easy interaction with organic pollutants. Gelatin limitations, including its hydrophobic nature and less mechanical stability, were overcome by forming composites with other desirable materials that showed unique features (Rigueto et al., 2021).

13. Adsorption and kinetic studies

Adsorption kinetics models play a crucial role in understanding the intricate details of the adsorption process, including the manner, mechanism and speed at which adsorption occurs in batch studies. Researchers aim to determine the adsorption capacity and identify the best isotherm models for the adsorption of drugs and dyes. Isotherms provide valuable insights into the relationship between contaminant concentration and adsorption at equilibrium, helping to identify optimal conditions and select suitable models. The correlation coefficient (R^2) is utilized to assess the fit between experimental data and different isotherm models, facilitating model selection (Osagie et al., 2021).

The Langmuir adsorption isotherm describes the relationship between the amount of adsorbate and its equilibrium concentration on an adsorbent in a water solution. It assumes a strong interaction between the adsorbent surface and the adsorbate, a specific number of available adsorption sites and the formation of a monolayer of adsorbed molecules. This equation aids in understanding and modeling the adsorption process between a solid surface and a solution (Ghaedi et al., 2012).

The Freundlich isotherm is an empirical equation widely used to describe the adsorption equilibrium of pollutants on biosorbents. While the Freundlich and Langmuir models can describe many biosorption isotherms, they do not consider external environmental factors. The complex nature of biosorbents and solution chemistry can lead to irregular patterns in biosorption isotherms, cautioning against mechanistic conclusions solely based on model fitting (Mohan and Singh, 2002; Wang and Chen, 2006).

The Brunauer-Emmett-Teller (BET) is an extension of the Langmuir theory, accounting for the formation of multiple layers of adsorbate molecules on the surface. It provides a better description of physical adsorption compared to the Langmuir isotherm, which mainly applies to chemical adsorption (Neolaka et al., 2020b).

The Temkin isotherm model considers the intermolecular interactions between the adsorbent and adsorbate, resulting in a homogeneous distribution of binding energies. It causes a linear decline in the heat of adsorption as the adsorbent surface coverage increases.

On the other hand, the Sips isotherm model combines features of the Langmuir and Freundlich models. It behaves similarly to the Freundlich isotherm at low adsorbate concentrations but exhibits characteristics of the Langmuir isotherm during multilayer adsorptions at high concentrations. The adsorption capacity determined by the Sips isotherm is considered more realistic as it accounts for both monolayer and multilayer adsorption processes (Neolaka et al., 2023; Ramadoss and

Subramaniam, 2018).

Sorption kinetics studies provide valuable insights into diffusion processes, adsorption rates, and rate-limiting steps involved in adsorption. Various models, such as pseudo-first-order (PFO), pseudo-second-order (PSO), Elovich, and the intraparticle diffusion model, have been employed to explain solute dispersion on the sorbent surface and within pores. The PFO model assumes a proportional relationship between the rate of change in adsorbed solute and the equilibrium adsorption capacity, while the PSO model suggests a chemisorption mechanism involving electron exchange or sharing between the sorbate and sorbent (Asl et al., 2013). The Elovich model describes heterogeneous diffusion processes involving reaction rates and diffusion factors, encompassing various reaction mechanisms. On the other hand, the intraparticle diffusion model focuses on the transportation of the adsorbate within the solid phase, determining whether the adsorption process is controlled by the adsorption rate (Wang et al., 2021).

Table S7 depicts some examples of biosorbents following different adsorption isotherms and kinetics model for uptake of pollutants. The mathematical formulae of the different adsorption isotherms and kinetic models are shown in Tables S8 and S9, respectively.

In a study investigating Cr(VI) adsorption using Cr(VI)-imprinted poly (4-VP-*co*-MMA) (IIP) supported on activated Indonesia natural zeolite, the maximum adsorption efficiency was observed at pH 2, with a 30 min contact time and at 30 °C. Kinetic and isotherm models were employed, revealing that the adsorption followed the PSO kinetic model and the Langmuir isotherm model, indicating a chemisorption process (Neolaka et al., 2018).

The blue-green algae *Spirulina platensis* was studied for the adsorption of acid blue 9 and FD&C red no. 40, showing maximum adsorption values of 1619 and 468 mg/g, respectively, at an optimum pH of 4. The Sips kinetic model provided the best fit for the experimental data (Dotto et al., 2012).

GO derived from kusambi wood was utilized to synthesize a magnetic nanocomposite adsorbent (GO-Fe_3O_4) for the efficient adsorption of Cr(VI) ions. Optimal conditions for adsorption were determined, and the kinetics followed a PSO model, while the isotherm fitted the Langmuir model. Experimental data of takari sand-derived silica@mercapto against Cu and Pb showed that the reaction followed pseudo-second-order kinetics. The high values of R^2 (0.969) for Cu(II) and 1.000 for Pb(II) indicated a strong correlation between the adsorbent and adsorbate, suggesting a proportional relationship between the adsorption capacity and the number of active sites (Neolaka et al., 2020a,b).

Mohan and Singh evaluated the use of modified bagasse for removing cadmium and zinc from wastewater. The Freundlich isotherm provided a more adequate fit than the Langmuir model, with higher adsorption capacities observed at 40 °C, indicating an endothermic adsorption process Mohan and Singh, 2002. An activated natural zeolite-magnetic composite material was synthesized and examined for the uptake of Cr ions. The adsorption mechanism followed the PSO model, with the Langmuir isotherm providing the best fit and a maximum adsorption capacity of 2.850 mg/g (Neolaka et al., 2022). A hydrochar derived from Bali cattle bone was effective in adsorbing methyl red from water samples. The adsorption followed a PSO kinetic model and adhered to the Freundlich isotherm, suggesting chemisorption with endothermic and spontaneous reactions (Neolaka et al., 2023).

14. Desorption/regeneration of plant based adsorbents

The ability to regenerate is a critical characteristic of adsorbents as it can pose significant challenges to achieve efficient adsorption rates, when the adsorbent loses its effectiveness after a few cycles. If an adsorbent loses its efficiency, the process of decontaminating water through adsorption can become quite expensive. However, the use of biogenic wastes as adsorbents can be more economical and can help reduce environmental waste. Once the dyes or other pollutants are desorbed from the adsorbent, it can be reused again. There are various processes, such as catalytic and chemical decomposition, solvent extraction, thermal treatment and radiation, that can enable the adsorbent to be recovered and reused (Ashiq et al., 2021).

Biomass-based adsorbents offer a unique advantage, as they can bypass the challenges associated with regeneration. They can be easily reused with or without modifications, due to their strong affinity with dyes, or they may not require significant regeneration at all. These adsorbents are abundant in nature, cost-effective and readily accessible in several places, being attractive alternatives to traditional adsorbents for water decontamination (Nayagam and Prasanna, 2021).

Various mechanisms, including precipitation, complexation, and ion exchange, can be utilized to recover inorganic and organic compounds from solutions. Regeneration agents, such as acids (HCl, H_2SO_4, HNO_3) and bases (NaOH, $NaHCO_3$, NaH_2PO_4), are commonly employed to desorb pollutants from biochar. Alkaline agents are particularly effective to desorb contaminants like arsenic (As) from Fe-impregnated biochar (He et al., 2018; Rosales et al., 2015).

Concerning desorbing pollutants from sorbent surfaces, different methods can be employed. Inorganic acids like HCl, HNO_3, and H_2SO_4 can recover metals through proton exchange. However, the use of these acids can potentially damage the biosorbent surface and reduce its capacity. On the other hand, organic acids, such as CH_3COOH and $C_6H_8O_7$, offer a more environmentally friendly and compatible alternative for desorption, as they do not harm the surface structure of the biosorbent (Albadarin et al., 2011; Jing et al., 2007).

The reusability of Bali cow bone hydrochar as an adsorbent was investigated using methyl red solutions. After four adsorption cycles, the material demonstrated a rejection efficiency of 86.9% and a desorption efficiency of 82.38%, highlighting its potential for treating methyl red effluent (Neolaka et al., 2023).

Ethanol was employed as an eluent for regenerating *Jacaranda mimosifolia*, after adsorption of ketoprofen. Over the course of 5 cycles, the removal percentage decreased from 89.45% to 81.59%, indicating a decay of 7.86%. Although the material exhibited an excellent regeneration response, it is worth noting that the disposal of the adsorbent becomes crucial as the number of cycles reduces its available sites (Georgin et al., 2021).

The efficacy of palm empty fruit biochar (PEF) for adsorbing Cibacron blue dye was investigated, and it exhibited a high adsorption capacity of 99% (Jabar and Odusote, 2020). In order to test the regeneration ability of PEF, seven consecutive adsorption-desorption cycles were conducted, which resulted in a small reduction in the adsorption capacity from 99% to 92.27% in the last regeneration cycle.

Vaccinium myrtillus (bilberry) leaves were used as an adsorbent to remove the cationic dye MB from water bodies. Several regenerating agents were employed to desorb the dyes from the adsorbent, and the desorption of the dye was calculated using the following equation:

$$D(\%) = m_d/m_a \times 100$$

where m_a represents the amount of dye adsorbed by the material, and m_d is the quantity of dye released by the regenerating agent (Mosoarca et al., 2022).

15. Limitations

Improving the limitations of bio-adsorbents for pollutant adsorption is an essential area of research to address the environmental concerns associated with their current methods of preparation and use. Some potential improvements and considerations include:

Green preparation methods: The limitations of bio-adsorbents for pollutants adsorption, include reliance on various chemical treatments and elevated temperatures for adsorbent preparation. It is important to explore more alternative, environmentally friendly methods for preparing bio-adsorbents that reduce or eliminate the reliance on chemical treatments and elevated temperatures. This could involve developing novel extraction techniques or optimizing existing methods to minimize the use of chemicals and energy.

Sustainable resource management: It is known that biogenic adsorbents derived from agricultural waste or other biomass are considered more environmentally compatible but have their own drawbacks. For example, the pre-processing of agricultural waste for production of activated has a notable environmental impact, including a significant contribution to ozone depletion. The activated carbon production from papaya peels has significant environmental impacts, contributions to global warming, ozone formation, ionization radiation, acidification, eutrophication, ecotoxicity, carcinogenic and non-carcinogenic toxicity, land use and water consumption (Aneja et al., 2009). Agricultural wastes have a significant environmental impact, contributing to 54.64% of ozone formation, 33.06% of fine particles, 27.86% of global warming and 98.24% of marine eutrophication (Noman et al., 2022). Thus, the development of innovative technologies remains crucial to address these limitations (Olguín and Sánchez-Galván, 2012). It is important to investigate the use of renewable resources for bio-adsorbent production. Instead of relying only on agricultural waste, explore other biomass sources that have lower environmental impacts or are considered waste from other industries.

Minimize chemical additives: Develop strategies to minimize the use of chemical additives during the adsorption process. This could involve exploring alternative pretreatment techniques or modifying the adsorbent structure to enhance its adsorption capacity without the need for additives. By reducing the reliance on chemical additives, potential risks associated with acid leachate containing toxic pollutants can also be mitigated. A study emphasizes the challenges associated with the use of chemical additives and the treatment of acid leachate containing toxic pollutants during adsorption process (Contescu et al., 2018). The use of phosphoric acid in the process majorly contributes to resource scarcity, particularly fossil resource scarcity (49.09%), while its impacts on terrestrial, marine, and freshwater ecosystems are relatively minimal. The environmental effects of inorganic acids, however, are not well-documented and require further research (Cruz et al., 2019).

Microbial optimization and genetic engineering: Biosorption also utilize microorganisms to remove different pollutants, involving passive and active transport mechanisms. The process is similar to adsorption with activated carbon but requires pretreatment of microbial cells for improved efficiency. Bioaccumulation within living cells offers great efficiency in reducing pollutant toxicity, but its commercial applicability is limited, requiring optimization of microbial growth conditions or the use of genetically engineered microorganisms (Sun et al., 2022). The microbial biomass used as adsorbent faces challenges in scaling up to industrial processes due to high production costs, technical complexity, and environmental risks. So it is important to focus on optimizing microbial growth conditions or modifying their genetic makeup of microorganisms, so that it may be possible to increase the effectiveness of bioaccumulation within living cells, thereby reducing pollutant toxicity more efficiently.

Cost reduction and scale-up: Investigate strategies to reduce the production costs associated with bio-adsorbents and develop scalable manufacturing processes. This could involve exploring new technologies, process optimization, or integrating bio-adsorption systems into existing industrial processes. Addressing the challenges related to high production costs, technical complexity, and environmental risks will be crucial for the successful implementation of bio-adsorbents in industrial applications.

Innovative technologies: Continue to invest in research and development of innovative technologies for pollutant adsorption. This could include exploring alternative materials, novel adsorption techniques, or hybrid approaches that combine bio-adsorbents with other advanced materials. By fostering innovation, scientists and engineers can find new solutions to overcome the limitations associated with current adsorption technologies.

It is important to note that these suggestions provide a starting point for addressing the above mentioned limitations. Further research and collaboration between scientists, engineers and policymakers will be necessary to develop practical and sustainable solutions for adsorption using bio-adsorbents.

16. Conclusions

This comprehensive review provides an overview of non-conventional biogenic adsorbents and their operating conditions for the effective removal of drugs and dyes. Conventional wastewater treatment processes generate secondary pollutants, which pose significant environmental challenges. To address this issue, low-cost adsorbents derived from plants, agricultural waste, microorganisms, animal waste, biopolymers, algae and biogenic minerals can be used, due to their appropriate surface functional groups, which attract pollutants.

Polysaccharides, such as cellulose, starch and CT exhibit unique characteristics, like excellent structural composition, abundance and ease of modification, making them promising adsorbents. Additionally, bacteria, fungi, yeast, and algae are highly efficient in adsorbing drugs and dyes, by providing access to their internal surface area through a relatively large porous network.

The use of these biogenic adsorbents offers a sustainable, eco-friendly and biodegradable solution for the remediation of dyes and drugs, which is of utmost importance for sustainability. By utilizing these low-cost adsorbents, environmental challenges can be addressed and a greener and cleaner future can be promoted, although further studies are required to overcome the limitations.

17. Future recommendations

This review highlighted naturally available adsorbents as biodegradable and cost-effective alternatives to commercially available activated carbon. Adsorbents derived from biogenic biomass or waste are comparatively considered as low-cost adsorbents. However, the emergence of complex and diverse aquatic dyes and drugs requires the development of multifunctional modified lignin-based adsorbents. In order to enhance the adsorption capacity, it is necessary to analyze the chemical and morphological structure of all biogenic adsorbents.

Although researchers are investigating biogenic biomass derivatives as adsorbents, large-scale applications are still limited. Conducting large-scale studies is crucial to fully leverage the advantages of these eco-friendly adsorbents and minimize the generation of secondary pollutants during water remediation processes. The existing literature provides valuable insights and guidance on optimizing reaction conditions, including the determination of optimal parameters, for effectively treating multiple drugs and dyes. In order to stabilize biogenic adsorbents and provide high surface-to-volume ratios and versatile functionalities, further immobilization of biopolymers or biosorbents by nanostructured materials is required. It is essential to develop strategies to recycle adsorbed dyes and drugs into organic or other commercially valuable products for the benefit of mankind.

Additionally, the compatibility of adsorbents with reaction conditions, such as pH, temperature and contact time must be ensured. By addressing these challenges, more efficient, cost-effective and environmentally friendly biogenic adsorbents can be developed for the removal of drugs and dyes from water bodies (see Figs. 11 and 12).

Author contributions

Mobeen Ur Rehman: writing - original draft, conception; Muhammad Babar Taj: conception, supervision, resources, project administration; Sónia A.C. Carabineiro: supervision; visualization; writing - reviewing and editing.

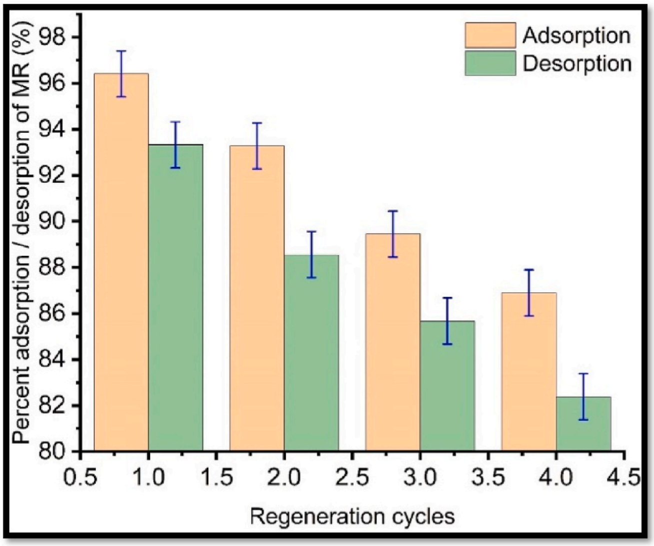


Fig. 11. Regeneration cycle of Bali cow bone hydrochar against methyl red. Reprinted from (Neolaka et al., 2023) with permission from Elsevier.

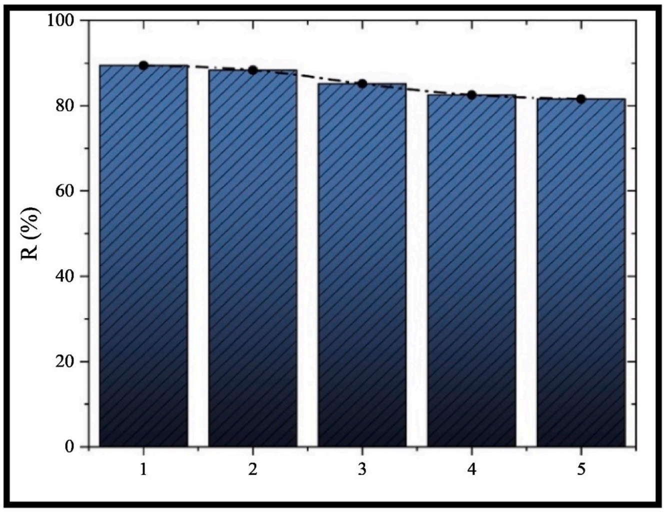


Fig. 12. Desorption studies of ketoprofen from Jacaranda mimosifolia. Reprinted from (Georgin et al., 2021) with permission from Elsevier.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

Acknowledgments

M.B.T acknowledges support from HEC (Higher Education Commission) of Pakistan. S.A.C.C. acknowledges support from FCT/MCTES (Fundação para a Ciência e Tecnologia and Ministério da Ciência, Tecnologia e Ensino Superior) through projects UIDB/50006/2020 and UIDP/50006/2020 and for the Scientific Employment Stimulus—Institutional Call (CEECINST/00102/2018).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2023.139477.

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