REVIEW

Modified biomass adsorbents for removal of organic pollutants: a review of batch and optimization studies

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Abstract

Modification of the adsorbent surfaces has been considered a fascinating strategy that enhances biomass-based adsorption properties for efficient removal of organic pollutants. This is based on the attempt to replace the cost-ineffectiveness of the commercial activated carbon. The present study discusses different modification strategies and a review on modified biomass materials for the sorption of organic contaminants. Unlike previous literatures in the field, wider range of these pollutants are discussed in this study under different categories including pesticides (such as insecticides, herbicides, fungicides), pharmaceutical (e.g. analgesic and antipyretic drugs, antibiotic drugs, non-steroidal anti-inflammatory drugs and antimalaria drugs), and dyes (e.g. azo, xanthene, miscellaneous diagnostic, tri-aryl methane, and phenol-derived polymeric dyes). It was observed that the acid-activated *Posidonia oceanica* and HNO_3-modified rice husk displayed the highest and lowest adsorption capacities of 2681.9 and 0.35 mg/g for removing Rhodamine B dye and methyl parathion pesticide, respectively. The mechanistic aspects of organic pollutants adsorption, their corresponding regeneration studies, and environmental challenges with chemical modifications are also discussed. The use of computational (optimization) models for modified biomass-based adsorbents to remove organic pollutants is devoid in previous reviews but discussed in the present study. To foster more advancement in this field, the concluding part presents various challenges and knowledge gaps for furthering research towards more realistic industrial implementations.

Abbreviations

ACs	Activated carbons
ACFs	Activated carbon fibres
EDA	Electron donor–acceptor
BSAC	Banana stalk activated carbon
CBS	Cob bio-waste sorbents
PUHs	Phenylurea herbicides
FAO	Food and Agricultural Organization
RSB	Raw straw biochar
IS	Ionic strength

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PVA	Polyvinyl alcohol
I.C	Initial conditions
NA	Not applicable
Ps1	Pseudo-first order
Ps2	Pseudo-second order
RSM	Response surface methodology
CCD	Central composite design
BBD	Box–Behnken design
MB	Methylene blue
PFPAC	Pomegranate fruit peel activated carbon
MG	Malachite green
AI	Artificial intelligence
ANN	Artificial neural network
NPs	Nanoparticles

Introduction

The advances in the industrial revolution and urbanization have resulted in day-to-day increase in manufacturing of chemical substances like pesticides, herbicides, pharmaceutical and personal care products, dyes, etc. with their respective ever rising and continuous demands for different applications (Philippot et al. 2015; Ahmad et al. 2019; Ibrahim et al. 2021; Adegoke et al. 2022c, a). Discharge of effluents from these manufacturing and applications industries has led to the discharge of hazardous compounds into the ecosystems (De Gisi et al. 2016; Bulgariu et al. 2019; Agboola and Bello 2020; Kassegne et al. 2020; Li et al. 2020; Ahmad

et al. 2021c; Bello et al. 2021). The accumulation and movement of same into water bodies via erosion, drought, etc. have become a global threat into both human and aquatic habitat (Fig. 1) (Adegoke and Bello 2015; Ahmad et al. 2016; De Gisi et al. 2016; Singh et al. 2017; Thambiraj et al. 2018; Bulgariu et al. 2019; Adebusuyi et al. 2022). Pollutants pose adverse effects on the ecosystem, thereby making the sustainable environment sustenance/maintenance practically impossible (Bello et al. 2015b; Bellec et al. 2015; Wang et al. 2016; Bello et al. 2017a; Adegoke et al. 2017, 2019; Jain and Gogate 2017b; Yaseen and Scholz 2018). Water is important to life and it is a major component of humanity, and other aquatic systems; therefore, water pollution resulting from climate change, industrialization, urbanization, and population is projected to result in possible potable water scarcity in the nearest future (Afolabi et al. 2020a; Ahmad et al. 2021c, b).

However, the rise in applications and conversion of biomass-based wastes into products with added value such as adsorbents and catalysts for different applications has been witnessed in recent years. Based on this, numerous adsorbents have been produced from biomass-based origin as alternative for the cost-ineffective commercial activated carbon which are not sustainable (Bello et al. 2015a, 2019a; Pathania et al. 2017). Also, this approach remains a fascinating research hotspot since it possesses the benefit of enhancing environmental management system by preventing biomass wastes' accumulation in the environment. This is because the accumulation of these wastes causes serious
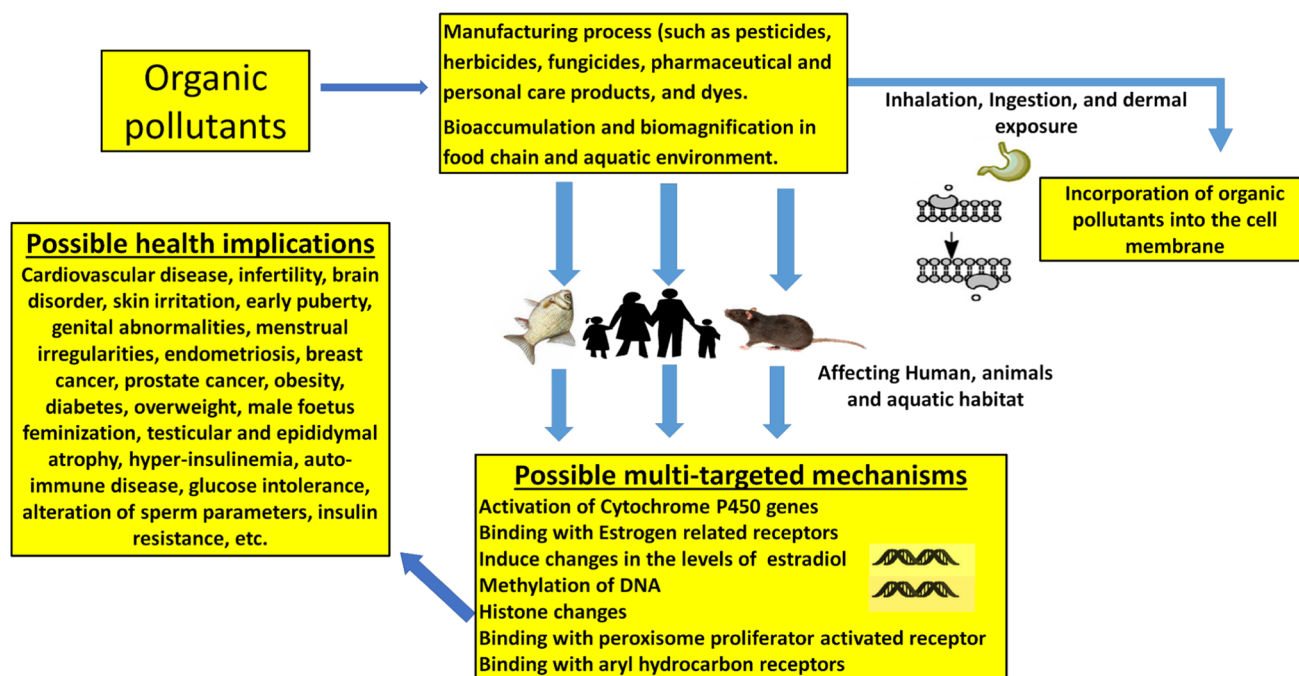

Fig. 1 Accumulation and movement of organic pollutants and their corresponding possible health implications

air, water, and soil pollutions. They become worse in rainy season, generate terrible odour and stink at the fall of rain. They are difficult to manage by environmentalists and usually end-up in streams, rivers and oceans thereby endangering the aquatic habitat. Burning these wastes even during dry season causes air pollution and the death of important microbes/biological systems (Bello et al. 2017d, 2020c, 2021; Ojedokun and Bello 2017a; Bello et al. 2019a, b, c; Yu et al. 2019; Afolabi et al. 2020b; Ahmad et al. 2021b, 2022). These abnormalities have inspired researchers worldwide to convert the biomass wastes into important products for water/wastewater treatment.

It has however been observed that raw adsorbents derived from biomass sources have poor adsorption capacity; therefore, various enhancing strategies have been employed to facilitate the sorption ability adsorbents for effective removal of contaminants (Khodaie et al. 2013; Fernandez et al. 2014; Utomo et al. 2015; Singh et al. 2017). One major fascinating strategy is surface modification of the adsorbent thereby resulting in both physical and chemical changes with structural enhancement needed for effective adsorption properties such as higher active sites, higher surface areas, enhanced particle and pores sizes, improved porosities, and pore volumes (Üner et al. 2017; Khasri et al. 2018; Sham and Notley 2018; Bello et al. 2019b, 2020a; Ahmad et al. 2020). For example, some recent studies conducted using modified biomass-based materials such as Dragon peel (Ahmad et al. 2021b), pod of *Mangifera indica* (Bello et al. 2021), lemon grass (Ahmad et al. 2019, 2021a, 2022; Putri et al. 2020), pomegranate fruit peel (Ahmad et al. 2020); leaves of *Gmelina aborea* (Bello et al. 2020a), husks of coconut (Bello et al. 2019a), *Parkia biglobosa* (Bello et al. 2019b), bean husk (Bello et al. 2017d), *Moringa oleifera* leaf (Bello et al. 2017b, c), guava leaf (Ojedokun and Bello 2017b), berry leaves (Ahmaruzzaman et al. 2015), *Glossogyne tenuifolia* leaves (Yang and Hong 2018), *Ficus racemosa* (Jain and Gogate 2017c, a), coconut leaves (Cazetta et al. 2011; Rashid et al. 2018), *C. camphora* leaves (Tang et al. 2017), leaves of plane trees (Gong et al. 2013), senescent plant leaves (Gunasekar and Ponnusami 2013), durian leaves (Hussin et al. 2015), and bamboo (Ghosh and Bandyopadhyay 2017), *Prunus dulcis* (Jain and Gogate 2017b) have been reported. They have reported how effective the strategy was for enhancing sorption properties of adsorbents for different organic pollutants removal. Thus, fabricating novel functionalized adsorbents for effective removal of different organic pollutants remain a global research hotspot.

Conversely, it should be noted that some studies have reported the utilization of numerous sorbents derived from clay, sand, nanoparticles, and other synthetic adsorbents. However, no study has presented the comprehensive information on the modified biomass-based adsorbent for organic pollutant removal focusing entirely on pesticides, e.g. organophosphate, oximino carbamate, atrazine and thioatrazine, etc.; insecticides, e.g. neoncotinoids, n-phenylurea, sulphometuron-methyl, etc.; fungicides, e.g. azoxystrobin; pharmaceutical pollutants, e.g. paracetamol (acetaminophen), ciprofloxacin, doxycycline, ibuprofen, lumefantrine, dicloxacillin, phenacetin, levofloxacin, hydrochlorothiazide, etc.; and dyes, e.g. Congo red, tartrazine, rhodamine B, methylene, phenol-derived polymeric dyes, indigo carmine, methyl red, acid green 3, azo dye, reactofix red 3BFN, direct blue 53, reactive red 4 dye, etc. discussed in this review. In addition, the mechanistic aspects of organic pollutants' adsorption and corresponding regeneration studies are also discussed. Unlike existing reviews in the field, the present study presents the review of various computational (optimization) models for the adsorption of organic pollutants using modified biomass-based adsorbents, covering response surface methodology and artificial intelligence. The concluding part of the work presents various challenges, and knowledge gaps for furthering research in the field towards more realistic industrial implementations.

Routes of exposure to organic pollutants and their ecotoxicological effects

Toxicity means ability of a substance to cause harmful effects in living organisms depending on its dose, route of entry, frequency and duration of exposure, environmental factors, chemical combinations, and inter- and intraspecies variations. There are three major routes by which organic pollutants can enter into the body system of living organisms: inhalation, absorption (skin or eye), and ingestion (Vafeiadi et al. 2014). Inhalation is the most common route of entry of organic pollutants into living organisms. The inhaled and deposited organic pollutants are bioaccumulated in fatty tissues of living organisms and may cause various health effects. Absorption of organic pollutants through the skin (dermal) is also a route of entry into the body of living organisms. Once organic pollutants are absorbed in large quantities by the skin, they may produce systemic damage to internal organs. The eyes can also absorb organic pollutants and transport them to other parts of the body thereby causing harm to the organism. Ingestion of organic pollutants is also very common through the consumption of foods that are contaminated with organic pollutants (Kassegne et al. 2020). These organic pollutants are absorbed by the lining of the gastrointestinal tract and then transported to internal organs where they cause harmful effects. Several organic pollutants

have endocrine disrupting properties (Schug et al. 2011), and many are considered to be carcinogenic in living organisms. Organic pollutants usually have different potencies, and this is further complicated by the fact that no environmental exposure to organic pollutants is by a single chemical but by a mixture of organic pollutants. Several environmental conditions can affect the toxicity of organic pollutants such as climatic condition, temperature, salinity, nutritional status, adaptation to climatic change, and pH.

Organic pollutants are chemical substances which possess enormous ability to adversely affect the endocrine system of living organisms, can be either synthetic or natural (Arthur et al. 2019). They are usually leached from agricultural lands and dump sites into water bodies via erosion (Kabir et al. 2015). These pollutants are also transported to soils around during infiltration and water percolation, which then seep into groundwater via water movement in due course. This means that environmental pollutants can enter into human body system via contaminated food, air, and water.

They tend to alter normal endocrine and coordination functions through their action on hormonal glands (Falconer et al. 2006; Jung et al. 2015; Wee and Aris 2017). The level of coordination of hormonal system is very high with a very high specificity such that slight alteration in their concentration and timing result in massive distortions in their physiological properties leading to adverse health effects (Bai et al. 2016; Tapia-Orozco et al. 2016; Tursi et al. 2018). This therefore has enormous adverse human health effects because they cause the impairment of soil and water bodies. This suggests that their effects are troublesome since the alteration in genetic programming during their development in the early stages pose serious and significant effects which may result in transgenerational inheritance diseases (Schug et al. 2011; Skinner 2011; Raja et al. 2022). Previous study has demonstrated an illustrative model suggesting that all endocrine systems are targeted by these organic pollutants and these organs including the hypothalamic pituitary adrenal axis, pancreas, thyroid gland, and reproductive organs are vulnerable to attack by these organic pollutants since they are known to influence brain functions and hormone-dependent metabolic systems (Raja et al. 2022). Based on these exposure routes to organic pollutants and the corresponding ecological effects in the ecosystem, there is a serious need to develop efficient methods and readily available materials for decontaminating these noxious pollutants and ensuring sustainable and safer environment.

Fabrication processes of activated carbon for removal of organic pollutants

The quest for the use of activated carbon for adsorption of pollutants have been attributed to their desirable properties such as having microporous and homogenous structure in addition to their relatively high surface area as well as abrasion and very low cost of production in comparison to commercial activated carbon. However, characteristics of an activated carbon are reliant on chemical nature of the precursor and the fabrication process, which comprises two stages of carbonization and activation. The carbonization stage is a pyrolysis process that leads to thermal decomposition of the biomass-based material at temperature below 800 °C in the presence of minimal or no oxygen. This leads to the breaking down of lignocellulose structure of the biomass-based material and production of charcoal (Adegoke et al. 2022b).

The activation stage is required subsequent to the carbonization stage in order to copiously develop the pore structure that enhances activated carbon's surface area, which expedite the capture and retention of substances. The activation stage is performed using either chemical or physical treatment method which leads to the generation of activated carbon (Adegoke et al. 2022b). These various treatment methods during their preparation are responsible for the alteration of shape and size of leaves used as adsorbent (Adegoke and Bello 2015; Yahya et al. 2015). Treatment process by physical means takes place mainly through activation and carbonization. The precursors must first be carbonized before activation using carbon (IV) oxide (CO_2) or steam. Whereas, in chemical treatments; precursor materials are first of all impregnated by "activating agent" prior to heating in an inert atmosphere (Yahya et al. 2015). Dissolution of cellulosic constituents present in leaves are achieved by activating agents thereby enhancing the formation of crosslinks (Adegoke and Bello 2015; Yahya et al. 2015). In comparison, advantages of chemical activation over physical activation process are numerous and these include high production yield, ability to generate well formed microporous sites, high surface area, low temperatures, involvement of only one step (González-García et al. 2013; Adegoke and Bello 2015; Yahya et al. 2015), and mineral matter contents reduction that is higher (Bello et al. 2017b). Nevertheless, some shortcomings of chemical method of activation such as necessities to wash the activated/carbonized adsorbents to get rid of impurities that are associated with activation agents and corrosive nature of activating reagents still exist (Adegoke and Bello 2015; Yahya et al. 2015).

Preparation techniques for adsorbents through functionalization/modification

Among the several modification agents available, H_3PO_4 and $ZnCl_2$ are most widely utilized for activation of lignocellulosic adsorbents (González-García 2018). By comparison, many authors have shown H_2SO_3, H_3PO_4, citric

acid, and NaOH (Hussin et al. 2015; Makeswari et al. 2016; Ghosh and Bandyopadhyay 2017; Jain and Gogate 2017a, 2017c; Tang et al. 2017; Khan et al. 2018) preference to ZnCl$_2$ for modification of leaf because they are eco-friendly in nature. They have the ability to produce mesopores with high volume and diameter, and high surface areas (Adegoke and Bello 2015; Bello et al. 2017b; Ojedokun and Bello 2017b). Also, activated carbon produced are usually suitable for used in food and pharmaceutical companies (Yahya et al. 2015). Therefore, the use of H$_3$PO$_4$ as modification agent offers advantages such as (i) easy recovery of carbon product during processing, (ii) high amount of activated carbon is produced, and (iii) does not pose any toxic effect (Adegoke and Bello 2015; Yahya et al. 2015; Bello et al. 2017b; Ojedokun and Bello 2017b).

The mechanism of reaction for each activating reagent is different, for example, ZnCl$_2$ enhances extraction of H$_2$O molecules from lignocellulosic materials' structures while H$_3$PO$_4$ chemically associates within lignocellulosic materials (Yahya et al. 2015). Surface chemistry modification of leaves is an emerging, efficient, and promising method for applications of carbon in many fields. Modification of leaves involves oxidation and subsequent grafting onto leaf surfaces new molecule(s) such as cyclodextrin and functional group(s) such as –NH$_2$ and -COOH. Various modification methods of achieving oxidation are by acid, alkaline, ozone, microwave, and plasma. Commonly used modification methods are briefly discussed as follows:

Functionalization/modification by acidic methods

Acidic modification methods are majorly employed for oxidizing the porous carbon surfaces in order to increase the acidic properties thereby removing the excess mineral substances and also enhance the hydrophilic ability of the adsorbent surfaces (Table 1) (Shen et al. 2010). In recent years, nitric and sulphuric acids have been the most used acids though many others were used for modifying leaf adsorbents (Jawad et al. 2016; Jain and Gogate 2017b; Khan et al. 2018). These authors have established that utilization of acid functional group(s) existed on the surface of carbon upon modification for efficient dye removal from wastewater and aqueous solutions due to their oxygen-functional group containing-proton-donors.

The effect of acidic modification of bamboo leaves using citric acid was investigated for methylene blue adsorption in aqueous solution (Ghosh and Bandyopadhyay 2017). They tested seven isotherm models and Temkin isotherm gave the best description for the adsorption process while pseudo-second-order kinetic model best described the adsorption. Highest percentage removal achieved was 99.97%, with adsorption capacity of 725 mg/g. Bello et al. reported H$_3$PO$_4$-modified *Moringa oleifera* leaf for adsorptive removal of reactive blue (RB) dye (Bello et al. 2017c). They observed that the adsorption process of RB red dye was best described by Langmuir and pseudo-second-order models with intraparticle diffusion observed as adsorption process. They also observed that the

Table 1 Effects of acid-treated adsorbents for removing organic pollutants

Adsorbent used	Acid Used	Organic pollutant adsorbed	Adsorption capacity observed	References
Cocos nucifera shell	H$_2$SO$_4$	Methylene blue	Maximum adsorption capacity was observed at 50.6 mg/g	Jawad et al. (2020)
Moringa oleifera seeds	H$_3$PO$_4$	Diclofenac	Oxygenated functionality was observed, and 82.8% removal rate was reported at 95.85 mg/g due to the treatment	Bagheri et al. (2020)
Ashe juniper	H$_2$SO$_4$	Methylene blue	Maximum adsorption capacity increases from 0.30 to 0.97 mg/m^2, and oxygenated functionality such as carbonyl and sulphonated functional groups were observed as the result of the treatment	Choi et al. (2019)
Sawdust material	H$_2$SO$_4$ (1% and 40%)	NA	Specific surface area increases from 1.360 m^2/g to 44.4 m^2/g (1% H$_2$SO$_4$) and 135 m^2/g (40% H$_2$SO$_4$)	Benyoucef et al. (2020)
Weeds	HNO$_3$	Methylene blue	Maximum adsorption capacity increase from 161.29 to 39.68 mg/g	Güzel et al. (2017)
Peanut hull	H$_2$SO$_4$	Methylene blue	161.3 mg/g maximum adsorption was reported due to the treatment	Özer et al. (2007)
Peanut shell	H$_2$SO$_4$	Methylene blue and tetracycline	Maximum adsorption at 1250 mg/g (methylene blue) and 303 mg/g (tetracycline) were observed	Islam et al. (2019)

adsorption means energy evaluated from Dubinin–Radush-kevich isotherms confirmed the involvements of physical adsorption having a capacity of 934.4 mg/g. Many other authors also reported remarkable adsorption capacities with acid treatment (Oyelude 2015; Ghosh and Bandyopadhyay 2017; Ojedokun and Bello 2017b; Yang and Hong 2018; Mahmoudi et al. 2020). They discussed the different dye-uptake capacities attained from modified leaf samples in relation to surface chemical property of each adsorbent. They observed that anionic dyes on the prepared adsorbent samples by thermal treatment under hydrogen flow at elevated temperatures were favoured on considering their dispersive and/or electrostatic interaction.

Critical observation from literature established that acidic modification of leaf activated carbons (ACs) enhanced the adsorption of dyes onto the leaves-modified counterpart owing to a significant surface chemistry change. This is because acidic modifications have affinity to remove OH group(s), thereby producing the O_2-containing functional group(s), as the amounts of these functional groups have a close relationship with the ability of leaf ACs to remove dye compounds. Yang et al. (2016) used acid-treated vermicompost-derived adsorbent for the removal of congo red and methylene blue dyes from aqueous solution and observed similar findings. Xing et al. (2007) prepared active solid from carbon materials via sulphonation and they reported an enhanced material due to high steam-to-carbon ratio attributed to the acid treatment. This was further confirmed with FTIR results that indicated sulphonated groups at 1032 cm^{-1} (Xing et al. 2007) and ether groups at 1151 cm^{-1} (Lai et al. 2010) in similar experiments. N_2 isotherm curves indicated microporosity due to acid treatment (Güzel et al. 2017).

Functionalization/modification by alkaline (basic) methods

Alkaline (basic) modifications of leaf ACs produce positive-charge surfaces that help adsorb the significant amounts of negatively charged species. The porous carbons having the basic surface characters/properties can be achieved by treating the leaf materials in inert hydrogen or ammonia atmosphere at higher temperature such as 400–900 °C (treatment of leaves with NH$_3$) or at 400–600 °C (treatment with amide, aromatic amines or protonated amides) leading to the creation of basic formations with sufficient nitrogen functionalities (Faria et al. 2004). It is noteworthy here that adsorbents with nitrogen-containing functional groups are endowed with basic properties having the ability to trigger the enhancement of porous carbons and acid molecular interaction, e.g. hydrogen bonding, dipole–dipole interaction and/or covalent bonding. Interestingly, it is highly expected that the hydroxyl ions (OH$^-$) have a strong affinity to react with leaf ACs surface functional groups under a basic condition thus making this modification

treatments advantageous to enhance the adsorption of dyes. Basic modifications on leaves can be achieved through nitric acid treatment, partial gasification of oxygen, urea impregnations followed by pyrolysis. Generally, one can easily conclude that OH$^-$ ions are expected to react with the leaf surface functional group under alkaline modification thereby leading to the abundance creation of positive charges on leaf surfaces of AC which is favourable to enhance the negative-charged species from H$_2$O.

Zheng et al. (2013) modified activated carbon using NaOH and reported that there was increase in pore volume concentration and surface area of the adsorbent and thus improved adsorption of organic pollutants. However, using similar chemical, Sadaf and Bhatti (2016) reported that organic pollutants, such as anionic dyes, might not be effectively removed by alkaline-modified adsorbent because of the perceived surface deprotonation functionality due to –OH and –COOH that could cause an electrostatic repulsion of these dyes.

Hayati and Mahmoodi (2012) examined utilization of NaOH in surface modification of activated carbon and subsequent use of the prepared modified activated carbon in the removal of dyes. Their results showed that NaOH was effective for the surface modification. Compared to ordinary activated carbon surface, their results indicated higher maximum adsorption capacity of 9.17 and 11.77 mg/g onto the surface-modified activated carbon for Acid Red 14 (AR14) and Acid Blue-92 (AB92), respectively.

Functionalization/modification by impregnation methods

The phenomenon called impregnation is defined as fine, even or uniform distribution of chemical reagents and particles in the pores of leaf ACs. ACs from leaves can be impregnated with metal compounds from silver, copper, aluminium, iron, etc. due to their adsorption capacities that is significantly high. It should be noted that effect of impregnation ratio must be considered. This is the weight ratio of the activation agents to that of the precursor. This ratio is the most important factor in the activation process by chemical method because, with the increasing ratio, the leaf adsorbent surface area is expected to increase. Bello et al. impregnated *Moringa oleifera* leaf with NaOH and H$_2$SO$_4$ and used different characterization techniques to establish the effects of modification by impregnation. The study concluded that impregnated *Moringa oleifera* leaf could be employed for dyes recovery in lieu of commercial activated carbon (Bello et al. 2017b).

Magnetic modification

Aryee et al. (2020) prepared magnetic-modified biomass for dye removal using co-precipitation and yielded a crystalline

mesoporous material that possess superparamagnetic characteristics. The results obtained show improved sorption capacity of 32.5 mg/g by the modified biomass towards methylene blue. Safarik and Safarikova (2010) observed a monolayer adsorption in decontamination of different dye types using a magnetic-modified adsorbent via Langmuir model. Chen et al. (2011) carried out biomass pyrolysis with $FeCl_3$ and $FeCl_2$ to derive a magnetic material that could be applied in the separation of solid–liquid mixtures. They prepared the magnetic biochar via chemical co-precipitation of iron hydroxides on orange peel powder and subsequent pyrolysis. Orange peel was used as material and magnetite as magnetic medium. They observed that the novel magnetic biochar derived through their modification exhibited an enhanced adsorption capacity (several times) for phosphate and organic contaminants than untreated biomass. Hence, they indicated that magnetic-modified adsorbents could be an extraordinary sorbent for contaminants removal. The synthesis of magnetic-modified biomass has been carried out by scientists using different kinds of methods with various biomass and magnetic medium.

Functionalization/modification by microwave methods

Modification of leaf ACs via microwave radiation has been given wide popularity nowadays owing to its heating capacity at molecular levels which results in quick thermal

and homogenous reactions (Khan et al. 2018). Unlike conventional heating methods, microwave heating/irradiations offer significant advantages: (i) microwave energy neatly heats the leaf materials from inside out, (ii) provision of rapid heating by microwave energy, (iii) no need of heating convection through fluids, (iv) process of heating can be controlled easily, (v) direct contacts between the microwave heating sources and the heated leaf materials cannot occur, (vi) capable of been operated at higher temperatures, (vii) it saves energy and time, (viii) increasing chemical reactivities, (ix) cost-effective (x) systems processing are also moderately portable, compact and easy to maintain (Khan et al. 2018). For instance Khan et al. (2018) used spent black tea leaves prepared using microwave-assisted method as effective low-costs and green adsorbent for adsorption of dye (Khan et al. 2018). Their results revealed that Langmuir model best described the adsorption with adsorption capacity of 242.72 mg/g at 25 °C.

Performance of different modified biomass-derived adsorbents for removing organic pollutants

There has been series of scientific reports on application of modified biomass-derived adsorbent for removing organic and emerging contaminants such as dyes, pesticides, and
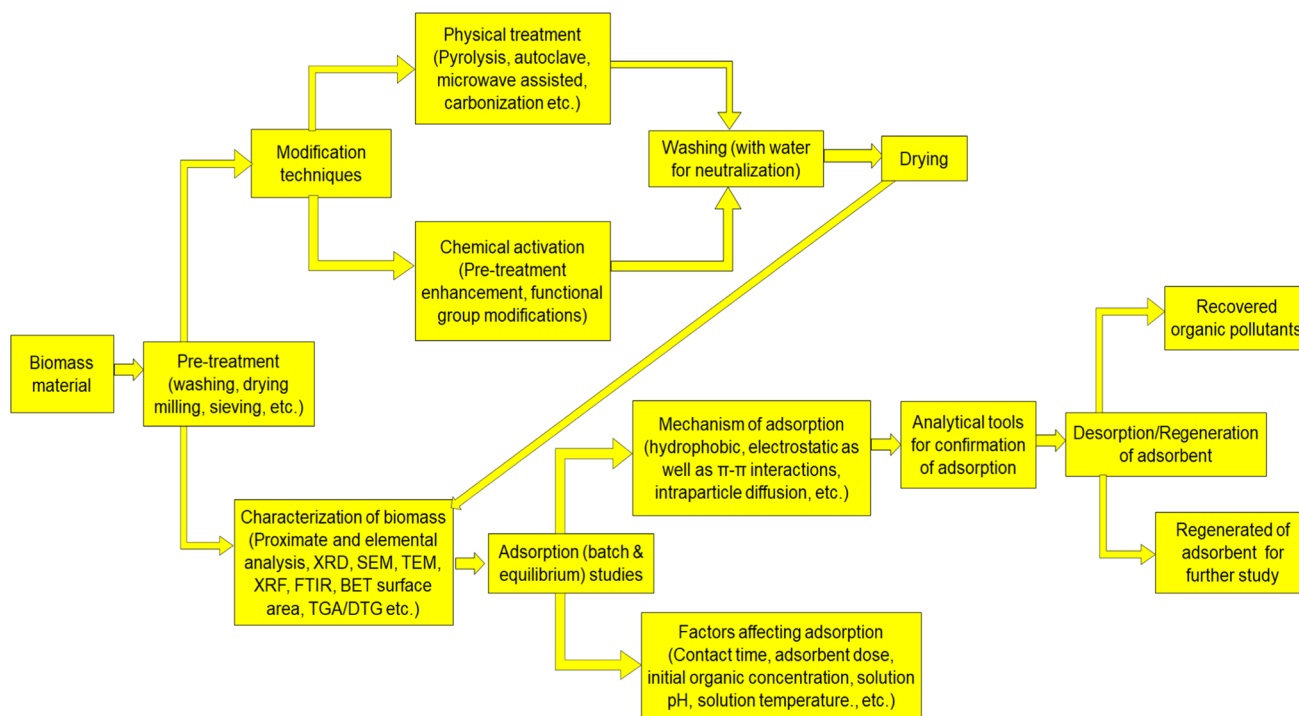

Fig. 2 Illustration of the overall adsorption process of organic pollutants onto modified biomass materials

Table 2 Modified biomass-derived adsorbents for organic pollutants removal

Adsorbents used	Pollutants removed	Initial operating conditions	Isotherm models	Kinetic models used	q_{max} (mg/g)	Adsorption mechanism	References
Modified cationic cellulose derived from coconut	Congo red	I. C = 40–100 mg/L, dose = 5–12 mg	L	Ps1	19.372	Chemisorption	Cocos (2021)
Modified cationic cellulose derived from coconut	Tartrazin	I.C = 40–100 mg/L, dose = 5–12 mg	F	Ps2	18.412	Chemisorption	Cocos (2021)
Ortho-phosphoric acid modified coconut husk	Rhodamine B dye	I.C = 200–1000 mg/L, dose = 0.1 g, T = 303–323 K	L	Ps2	1666.67	NA	Bello et al. (2019a)
H_2SO_4 modified coconut shell	Methylene blue	I.C = 25–200 mg/L, dose = 0.02–0.2 g, pH = 3–10	F	Ps2	50.6	Electrostatic attraction, hydrogen bonding, π interaction	Jawad et al. (2020)
Acid activated *Posidonia oceanica* (sea grass)	Methylene blue	Dose = 5 g, pH = 2–12, t = 10–90 min, I.C = 2–500 mg/L	D-R	Ps2	2681.9	Physisorption	Elmorsi et al. (2019)
Thermal-treated rice husk	Phenol	Dose = 10 g, pH = 5.8, I.C = 100 mg/L, dose = 0.5 g	NA	NA	NA	NA	Daffalla et al. (2020)
Ca(OH)$_2$-treated rice husk	Phenol	Dose = 10 g, pH = 5.8, I.C = 100 mg/L, dose = 0.5 g	NA	NA	NA	NA	Daffalla et al. (2020)
Acid/thermal-treated rice husk	Methylene blue	2.0 g, pH = 5–9, I.C = 0.1–0.5 mg/L	L	Ps2	103.11	NA	Moeinian and Mehdinia (2019)
NaOH-treated Rice husk	Paracetamol	pH (2.0–3.5), dose = 0.025–0.45 g, I.C = 50–110 mg/L, t = 0–150 min	L	Ps2	20.964	Physisorption-chemisorption	George Nche et al. (2017)
Rice straw biomass	Doxycycline	pH = 2–12, dose = 0.5–4.5 g, t = 10–70 min	F	Ps2 Ps2	432.90 131.58	Hydrogen bonding and π–π interaction	Zeng et al. (2018)
Rice straw biomass	Ciprofloxacin	pH = 2–12, dose = 0.5–4.5 g, t = 10–70 min	F	Ps2	131.58	Hydrogen bonding and π–π interaction	Zeng et al. (2018)
Acetic acid/hydrogen peroxide-treated rice husk	Indigo carmine dye	pH = 2–12, dose = 0.5–3.5 g, t = 10–60 min, 40–60 °C, I.C = 1×10^{-4}–9.0×10^{-4} mg/L	L	Ps1	9.275	NA	Jain et al. (2006b)
KOH-activated pomegranate fruit peels	Methylene blue dye	Dose = 0.1 g, T = 303–313 K, I.C = 25–300 mg/L, t = 0–1440 min	F	Ps1	235.58	Physisorption	Ahmad et al. (2021c)
Functionalized bean husk	Ibuprofen	Dose = 0.1 g, pH = 4.75, I.C = 10–50 mg/L, T = 303, 313, 323 K, t = 2–70 min	L	Ps2	50.0	Physisorption	Bello et al. (2019c)
Functionalized locust bean pod	Rhodamine B dye	Dose = 0.1 g, pH = 4.75, I.C = 200–1000 mg/L, T = 303, 313, 323 K	L	Ps2	1111.1	Physisorption	Bello et al. (2019b)

Table 2 (continued)

Adsorbents used	Pollutants removed	Initial operating conditions	Isotherm models	Kinetic models used	q_{max} (mg/g)	Adsorption mechanism	References
Functionalized *Gmelina aborea* leaf	Rhodamine B dye	Dose=0.1 g, pH=4.75, I.C=200–1000 mg/L, T=303, 313, 323 K	L	Ps2	1000	Physisorption	Bello et al. (2020a)
Functionalized lemon grass	Methylene blue dye	Dose=0.1 g, pH=2–12, I.C=25–500 mg/L, T=30–60 °C	K-C	Ps1	342.9	Physisorption	Ahmad et al. (2021a)
KOH modified mango (*Mangifera indica*) pod	Rhodamine B dye	Dose=0.1 g, pH=2–11, I.C=200–1000 mg/L, t==0–120 min	F	Ps2	500	Boundary layer intraparticle diffusion	Bello et al. (2021)
Chemically activated banana stalk	Lumefantrine	Dose=0.1 g, pH=3–11, I.C=20–100 mg/L, t=0–200 min	L	Ps2	102.1	Boundary layer intraparticle diffusion	Agboola et al. (2021)
Modified orange peels	Ibuprofen	Dose=0.1 g, I.C=5–25 mg/L, T=30–50 °C	L	Ps2	49.30	Physisorption	Bello et al. (2020b)
HCl-modified *Azolla filiculoides*	Acid Green 3 dye	Dose=0.5–7 g/L, pH=3–10, I.C=10–500 mg/L, T=30, 40 °C	L	Ps2	37.5	NA	Balarak et al. (2016)
PVA coated activated carbon from *Crotolariaburhia* and *Opuntia dellinii*	Azo dye	Dose=0.5–7 g/L, pH=3–12, I.C=10–40 mg/L, t=15–120 min	F	Ps1	19.23	NA	Gehlot et al. (2011)
PVA coated activated carbon from *Crotolariaburhia* and *Opuntia dellinii*	Azo dye	Dose=0.5–7 g/L, pH=3–12, I.C=10–40 mg/L, t=15–120 min	F	Ps1	19.23	NA	Gehlot et al. (2009)
Modified *Moringa oleifera* seed pod	Paracetamol	Dose=0.1 g, pH=2–11, I.C=10–50 mg/L, t=0–300 min	L	Ps2	20.284	Chemisorption	Ogunmodede et al. (2021)
KOH-activated Pomegranate fruit peels	Ciprofloxacin	Dose=0.05 g, I.C=50–300 mg/L, t=0–1440 min	F	NA	2.353	Ion exchange electrostatic repulsion	Elhag Elhussien (2017)
NH_4Cl Activated Pomegranate fruit peels	Acetaminophen	Dose=0.1–2 g, T=10–40 °C, I.C=100–500 mg/L, pH=2–9	L	Ps2	233	NA	Mashayekh-Salehi and Moussavi (2016)
H_2SO_4 modified Indian almond leaf	Dichloxacillin	Dose=0.05–0.3 g, pH=2–8, I.C=25–500 mg/L, t=0–3120 min	L	Ps2	71.04	Hydrogen bond van der Waal	Sunsandee et al. (2020)
H_3PO_4-modified Ayous sawdust	Phenacetin	Dose=0.5 g, pH=2–8, I.C=25–500 mg/L, t=0–300 min	F	Ps2	5.33	π–π interaction	Ngakou et al. (2019)

Table 2 (continued)

Adsorbents used	Pollutants removed	Initial operating conditions	Isotherm models	Kinetic models used	q_{max} (mg/g)	Adsorption mechanism	References
H$_3$PO$_4$-modified Cucurbitaceae peelings	Phenacetin	Dose = 0.5 g, pH = 2–8, I.C = 25–500 mg/L, t = 0–300 min	L	Ps2	15.43	π–π interaction	Ngakou et al. (2019)
Thermally/Microwave-activated Tea waste	Acetaminophen	Dose = 1–2 g, pH = 3–8, I.C = 20–100 mg/L, t = 0–120 min	L	Ps2	195.95	NA	Dutta et al. (2015)
Tea waste biomass	Ciprofloxacin	Dose = 0.2 g, pH = 4–10, I.C = 150–500 mg/L, t = 0–24 h, T = 30–60 °C	L	Ps2	238.10	Hydrogen bonding, electrostatic as well as π–π interaction	Li et al. (2018b)
Bamboo charcoal	Ciprofloxacin	Dose = 0.1 g, pH = 2.5–9.5, I.C = 0.7–70 mg/L,	L	Ps2	36.02	Ion exchange and hydrogen bonding	Wang et al. (2017)
Activated *Astragalus mongholicus* residue	Ciprofloxacin	Dose = 0.025–0.5 g, pH = 3–10, I.C = 10–300 mg/L	L	Ps2	43.668	Hydrogen bond electrostatic and π–π interaction	Shang et al. (2016)
H$_2$O$_2$-modified wheat husk	Reactofix Red 3BFN	pH 2.0–12, dose 4.0–24.0	L	NA	80.37	NA	Jain et al. (2006a)
H$_2$SO$_4$-modified sugarcane bagasse	Levofloxacin	Dose 0.02–0.1 g, I.C = 2–10 mg/L, t = 15–120 min	L	NA	0.847	NA	Khan et al. (2012)
H$_2$SO$_4$-modified sugarcane bagasse	Hydrochlorothiazide	Dose 0.02–0.1 g, I.C = 2–10 mg/L, t = 15–120 min	L	NA	1.396	NA	Khan et al. (2012)
H$_2$SO$_4$-modified sugarcane bagasse	Paracetamol	Dose 0.02–0.1 g, I.C = 2–10 mg/L, t = 15–120 min	L	NA	2.838	NA	Khan et al. (2012)
NaOH-modified sugarcane bagasse	Levofloxacin	Dose = 0.02–0.1 g, I.C = 2–10 mg/L, t = 15–120 min	L	NA	2.0465	NA	Khan et al. (2012)
NaOH-modified sugarcane bagasse	Hydrochlorothiazide	Dose = 0.02–0.1 g, I.C = 2–10 mg/L, t = 15–120 min	L	NA	3.655	NA	Khan et al. (2012)
NaOH-modified sugarcane bagasse	Paracetamol	Dose = 0.02–0.1 g, I.C = 2–10 mg/L, t = 15–120 min	L	NA	1.604	NA	Khan et al. (2012)
Urea-modified sugarcane bagasse	Levofloxacin	Dose = 0.02–0.1 g, I.C = 2–10 mg/L, t = 15–120 min	L	NA	1.8742	NA	Khan et al. (2012)
Urea-modified sugarcane bagasse	Hydrochlorothiazide	Dose = 0.02–0.1 g, I.C = 2–10 mg/L, t = 15–120 min	L	NA	4.117	NA	Khan et al. (2012)

Table 2 (continued)

Adsorbents used	Pollutants removed	Initial operating conditions	Isotherm models	Kinetic models used	q_{max} (mg/g)	Adsorption mechanism	References
Urea-modified sugarcane bagasse	Paracetamol	Dose = 0.02–0.1 g, I.C = 2–10 mg/L, t = 15–120 min	L	NA	3.096	NA	Khan et al. (2012)
H_3PO_4-modified *Euphorbia antiquorun L*	Direct Blue 53	I.C = 25–75 mg/L, flow rate = 5.0–15.0 mL/min, bed height = 5.0–10.0 cm	Y–NM	NA	142.35	NA	Sivakumar and Palanisamy (2009)
H_3PO_4-modified *Euphorbia antiquorun L*	Reactive Red 4 dye	I.C = 25–75 mg/L, flow rate = 5.0–15.0 mL/min, bed height = 5.0–10.0 cm	Y–NM	NA	131.63	NA	Sivakumar and Palanisamy (2009)
H_3PO_4-modified akpi shells	Imidacloprid	Dose = 0.02–0.1 g, I.C = 2–10 mg/L, t = 15–120 min	L	Ps2	43.48	Chemisorption	Urbain et al. (2017)
H_3PO_4-modified Silk worm faeces	Oxamyl	pH = 2–10, t = 5–180 min, dose = 0.1 g, t = 0–3 h, I.C = 10–20 mg/L	F	Ps2	625.00	Chemisorption	Mohammad and Ahmed (2017)
Cow bone char	Hexazinone, sulphometuron-methyl, ametryn, diuron, azoxystrobin	Dose = 0.01, 0.1, 1 g, t = 24 h, T = 25 °C, I.C = 5 mg/L	NA	NA	NA	π–π interaction	Mendes et al. (2017)
HNO_3 modified rice bran	Methyl parathion	Dose = 0.025–0.8 g, t = 10–210 min, T = 283–323 K, I.C = 0.3–3.8 × 10^{-3} mg/L, pH = 1–10	L	Ps1	0.39	Chemisorption	Akhtar et al. (2007)
HNO_3-modified rice husk	Methyl parathion	Dose = 0.025–0.8 g, t = 10–210 min, T = 283–323 K, I.C = 0.3–3.8 × 10^{-3} mg/L, pH = 1–10	L	Ps1	0.35	Chemisorption	Akhtar et al. (2007)
HNO_3-modified *Moringa oleifera* pod	Methyl parathion	Dose = 0.025–0.8 g, t = 10–210 min, T = 283–323 K, I.C = 0.3–3.8 × 10^{-3} mg/L, pH = 1–10	L	Ps1	0.39	Chemisorption	Ahmad et al. (2010) and Akhtar et al. (2007)
HNO_3-modified sugar cane bagasse	Methyl parathion	Dose = 0.025–0.8 g, t = 10–210 min, T = 283–323 K, I.C = 0.3–3.8 × 10^{-3} mg/L, pH = 1–10	L	Ps1	0.36	Chemisorption	Akhtar et al. (2007)

I.C Initial concentration, *t* Time, *dose* Adsorbent dosage, *T* Temperature, *F* Freundlich, *L* Langmuir, *T* Temkin, *Ps1* Pseudo-first order, *Ps2* Pseudo-second order, *E* Elovich, *DR* Dubinin–Radushkevich model, *K–C* Koble–Corrigan model, *TM* Thomas model, *Y–NM* Yoon–Nelson model, *NA* Not applicable

pharmaceuticals from water/wastewater as discussed below. The corresponding adsorptive removal process (Fig. 2) and performances at different operating conditions, kinetics, isotherms and their mechanisms are summarized in Table 2.

Modified biomass-derived adsorbents for pesticides removal

Insecticides

The biomass-derived adsorbent from spent tea leaves has been reportedly used in removing organophosphate pesticides such as dimethoate and chlorpyrifos in a concentration array of 20 to 100 mg/L. The spent leaves derived biomass adsorbent displayed a mean percentage removal of 42.11 and 72.98% for dimethoate and chlorpyrifos pesticides, respectively (Beheary et al. 2018). Furthermore, some studies about the feasibility of modified rice bran, rice husk, *Moringa oleifera* pod, and sugar can bagasse for removing organophosphate pesticides from aqueous solution at varied operating adsorptive conditions have been reported. This involves modification of these biomasses with HNO_3 and the utilization of the resulting adsorbents for removing methyl parathion. These adsorbents demonstrated adsorption capacity of 28 m^2/g (rice bran), 17 m^2/g (rice husk), 27 m^2/g (*Moringa oleifera* pod) and 25 m^2/g (sugar cane bagasse) for methyl parathion removal (Akhtar et al. 2007; Ahmad et al. 2010). The recounted adsorption percentage removal and optimum adsorption capacity at optimum experimental conditions for removing methyl parathion were 96.31% and 0.196 mg/g, respectively. Langmuir model gave the best description of the adsorption isotherm with respective maximum adsorption capacities of 0.39 mg/g (rice bran), 0.35 mg/g (rice husk), 0.39 mg/g (*Moringa oleifera* pod) and 0.36 mg/g (sugar cane bagasse) (Akhtar et al. 2007; Ahmad et al. 2010). Application of the same adsorbents for the adsorption of methyl parathion from wastewater samples gave percentage removal of 99, 97, 98 and 99% for rice bran, rice husk, *Moringa oleifera* pod and sugar cane bagasse, respectively. The studies on thermodynamics displayed an exothermic, spontaneous, and reaction that was feasible with a low entropy. On the other hand, the adsorption energy deduced from Dubinin–Radushkevich model ranging from 10.1 to 11.6 kJ/mol, which indicates a chemical adsorption mechanism (Akhtar et al. 2007).

More recently, adsorption of chloropyrifos, dimethoate, and malathion were studied by Jocić and co-workers on viscose fibre-derived activated carbon. In the study, dimethoate gave the least adsorption efficiency followed by malathion, while chlorpyrifos gave the most efficient adsorption. The material properties were observed to correlate with the uptake of these organophosphorus pesticides. It was concluded that activated carbon fibre was optimally efficient for the adsorptive removal of chlorpyrifos, with 240 mg/g as experimentally observed adsorption capacitances (Jocić et al. 2022).

The modification of *Ricinodendron heudelotii* shells (akpi shells) with ortho-phosphoric acid to prepare microporous activated carbon with enormous surface area of 11,179 m^2/g has been reported. The use of ortho-phosphoric acid to modify the akpi shell denotes a positive influence in adsorption properties for effective adsorptive removal of neoncotinoids such as imidacloprid from an aqueous solution (Urbain et al. 2017). A maximum imidacloprid adsorption removal was reported to be 90% while the adsorption capacity of 43.48 mg/g was observed. Langmuir isotherm model fitted well with R^2 value of 0.990 and pseudo-second-order model with R^2 of 0.998 with a chemisorption mechanism approach (Urbain et al. 2017).

The biomass-derived from spent tea leaves has been reportedly used in removing imidacloprid in a concentration array of 20 to 100 mg/L, demonstrating a mean percentage removal of 93.01% (Beheary et al. 2018). More recently, corn stalk and ZIF-67 were used to prepare a novel magnetic porous carbon which was prepared via an in-suit process followed by pyrolysis at high-temperature and acid picking, and used for adsorptive removal of thiamethoxam and imidacloprid in aqueous solution (Yang et al. 2022). The prepared ZIF-67/CS@C exhibited efficient absorption capacities for thiamethoxam and imidacloprid in water with maximum adsorption capacities of 133 and 189 mg/g, respectively. The efficiencies of removal for thiamethoxam and imidacloprid reached peak values of 99.13 and 99.65%, respectively. Further probe into the mechanism of adsorption revealed that pore filling, H-bond, and π-π electron donor–acceptor interaction might be main driving forces for the adsorption by ZIF-67/CS@C, and the reasonable pore size distribution and high external surface area was attributed to why the ZIF-67/CS@C showed better adsorption performance.

The experimental study on the utilization of H_3PO_4 to modify silkworm faeces in preparing environmentally friendly activated carbon has reportedly yielded an adsorbent with desirable properties. These properties include respective mean pore diameter and specific surface area of 0.2035 and 75.219 cm^2/g for the adsorption removal of oxamyl, which is regarded as an oximino carbamate pesticide (Mohammad and Ahmed 2017). Furthermore, it was suggested that the equilibrium removal of oxamyl on modified silkworm faeces in the adsorption process was attained at contact time of 120 min. Freundlich model with R^2 of 0.9975 gives the best interpretation for the adsorption isotherm when compared with Langmuir and Temkin models with respective R^2 of 0.9217 and 0.9163.

Salman and Hameed prepared banana stalk activated carbon for sorption of carbofuran insecticide from aqueous media at varied contact time, temperature, initial concentration, and pH. From the adsorption study, equilibrium sorption of carbofuran was observed to decrease slightly from 65.33 to 63.54 mg/g when the initial pH of the aqueous media was varied from 2 to 12. This was attributed to presence of excess H^+ ions which was considered to accelerate the adsorption of carbofuran with the anion OH^- in the aqueous solution (Salman and Hameed 2010). El-Geundi and co-workers observed that activated carbon prepared from cotton stalks was an efficient adsorbent for the adsorption of methomyl which is an insecticide from aqueous media. In the study, phosphoric acid-activation surface area of the activated carbon was relatively high (1600 m^2/g), equilibrium adsorption was achieved in 2.5 h and the sorption capacity of the activated carbon increased with increasing methomyl concentration and contact time; however, it decreased with increasing temperature as well as activation energy of sorption was reported as -2.35 kJ/mol suggesting that the sorption process was an exothermic reaction (El-Geundi et al. 2013).

Herbicides

Mandala and co-workers prepared biochars using eucalyptus bark, bamboo chips, corn cob, rice husk and rice straw for adsorption of atrazine from aqueous solution. They went further to prepare acid-treated rice straw and among these unreacted biochars, it was rice straw that showed the maximum adsorption for atrazine (70.7%) (Mandal et al. 2017). It was also noted that phosphoric acid treatment of rice straw improved its sorption. Besides, adsorption of atrazine on rice straw was best explained using the pseudo-second-order model. It was noted that the sorption decreases with increasing concentration of the atrazine in solution.

Recently, Phan et al. (2022) prepared various hydrochar using microwave-assisted hydrothermal carbonization under various conditions such as temperature ranging from 150 to 200 °C, residence time (20–60 min), and liquid to solid ratio from 5:1 to 15:1 mL/g as alternative adsorbent for atrazine removal. They activated the surface of hydrochar using different concentrations of H_2O_2 and KOH. Their results showed that as-prepared hydrochar at higher temperatures, longer residence times, and lower liquid to solid ratios showed significantly higher adsorption capacities for atrazine. From the results, KOH-activated hydrochar was reported to have highest adsorption capacity (4.06 mg/g) and even higher than biochar with higher surface areas. Other similar studies include hydrochar prepared by hydrothermal treatment of Prunus serrulata bark which is a novel and effective absorbent to adsorb atrazine in river waters with the maximum sorption capacity of 63.35 mg/g (Netto et al.

2022), while that of atrazine by HCl-modified corncob bio-waste sorbents at different pyrolysis temperatures and residence times revealed maximum adsorption in the range of 11.31–19.58 mg/g (Binh et al. 2022).

Recently, adsorption mechanism of rice straw biochar (RSB) to phenylurea herbicides (Monuron, Diuron, and Linuron) was studied via batch sorption experiments with three factors influencing the mechanism which are as follows: RSB dosage, ionic strength (IS), and pH, using orthogonal test (Dan et al. 2021). At pH value at 3, RSB dosage of 60 mg, and IS of 0.1 M, which was the optimal conditions for sorption, the maximum rate of removal attained for Diuron, Monuron, and Linuron were 25%, 41.9%, and 56.8%, respectively. Cara and co-workers investigated adsorption of sulphonylurea (chlorsulphuron) from an aqueous solutions using alkaline-treated wheat and corn (straw) mixed with soil and reported 337 mg/g maximum adsorption capacity for treated wheat and 318 mg/g for treated corn (Cara et al. 2017). Also, results obtained showed that maximum adsorption capacity in alkaline-treated straw was higher than in the soil (166 mg/g). Factors like the quality of raw material, hydrogen bonding interaction between chlorsulphuron and polar groups on the surface of the straw, π-π interactions between electron donor and acceptor, and hydrophobic moieties from the straw surface affect the adsorption capacity. Therefore, they concluded that the alkaline-treated straw has biosorption properties suitable for sulphonylurea removal. In another recent study, sulphuric acid was used to treat wheat husks (*Fagopyrum esculentum*) for its modification and applied as adsorbent for the removal of 2,4-dichlorophenoxyacetic acid (2,4-D) pesticide from aqueous solutions (Franco et al. 2021). Their findings showed that the maximum adsorption capacity attained was 161.1 mg/g at 298 K and electrostatic interactions was involved in adsorption mechanism.

Another novel study on the use of oxygen-defective graphdiyne as an material for removing the pesticides from water under various conditions showed remarkable sorption properties for seven sulphonylureas (Zhu et al. 2022). The maximum sorption capacity of oxygen-defective graphdiyne was 795.51 mg/g for iodosulphuron-methyl sodium, which was 130 folds higher than that of modified- graphene oxides and biochar with 6 and 1.5 mg/g respectively. The Langmuir isotherm model and pseudo-second-order kinetic model showed more suitability in describing this adsorption process, revealing that sulphonylureas onto oxygen-defective graphdiyne was monolayer coverage.

Fungicides

Adsorbent gotten from cow bone exhibited characteristic surface pore volume of 0.225 cm^3/g as well as surface area of 200 m^2/g. They have been apparently utilized as adsorbent

at various doses from 0.01 to 1 g in removing azoxystrobin fungicide that belongs to the class of β-methoxyacrylates pesticidal compounds (Mendes et al. 2017). It was stated that 0.01 g dose of cow bone char displayed azoxystrobin fungicide adsorptive removal from contaminated water. However, an increase in adsorptive azoxystrobin fungicide removal was proposed to increase with a consistent increase in the adsorbent doses of the cow bone char from 0.01 to 1 g. Furthermore, the mechanism of adsorption was proposed to occur by chemical interaction among functional groups on pesticides and the cow bone char which resulted in π–π interaction from the heterocyclic ring π electron donor (Mendes et al. 2017).

Modified biomass-derived adsorbents for pharmaceutical pollutants removal

Analgesic and antipyretic drugs

The utilization of NaOH has been presented to modify rice husk for removing paracetamol at varied operating parameters of pH (2.0–3.5), dose of adsorbent (0.025–0.45 g), initial concentration (50–110 mg/L) and contact time (0–150 min) in aqueous solution (George Nche et al. 2017). NaOH-modified rice husk exhibited 20.964 mg/g adsorption capacity for paracetamol as deduced from Langmuir isotherm with R^2 of 0.951. The kinetics of adsorption was best fitted into pseudo-second-order and intraparticle diffusion model with both having R^2 of 0.976, and the adsorption process was suggested to proceed in a competitive mechanism between physisorption and chemisorption (George Nche et al. 2017).

Chemically modified *Moringa oleifera* seed pods have been used in sorption of paracetamol from aqueous solution and the effects of operating experimental factors including initial concentration of paracetamol (10–50 mg/L), dose of adsorbent (0.1 g), contact time (0–300 min), pH (2–11) along with temperature (303–323 K) were considered (Ogunmodede et al. 2021). They observed that chemically modified *Moringa oleifera* showed a sorption capacity of 20.284 mg/g and Langmuir isotherm gave the best description of the sorption with $R^2 = 0.9947$ (Ogunmodede et al. 2021). The sorption kinetics was best fitted with the pseudo-second order with $R^2 = 0.9998$, while sorption removal of paracetamol was observed to proceed through the chemisorption process. Furthermore, thermodynamic study revealed that the removal of paracetamol using modified *Moringa oleifera* resulted in spontaneous, feasible, and endothermic chemical process. The cost analysis revealed that the chemically modified *Moringa oleifera* was about 10 times less expensive when compared to the commercial activated carbon premised on their respective cost price of 40.17 USD per kg and 398.79 USD per kg (Ogunmodede et al. 2021).

Modification of sugarcane bagasse using H_2SO_4, NaOH and urea as economical adsorbent for removing paracetamol from aqueous solution at various adsorption experimental operating conditions of initial drug concentration (2–10 mg/L), adsorbent dose (0.02–0.1 g) and contact time (15–120 min) has been studied (Khan et al. 2012). It was reported that the adsorptive removal of paracetamol increased proportionally with increasing initial paracetamol drug concentration from 2 to 8 mg/L, beyond which there was no noticeable significant increase in the adsorptive removal of paracetamol. Likewise, at optimal concentration of 8 ppm and varied adsorbent dose, a linear response in the relationship amid quantity of paracetamol drug adsorbed and dose of the modified sugarcane bagasse was observed. This connotes that adsorption capacity of the adsorbent increased with increase in the adsorbent dose (Khan et al. 2012). In addition, Langmuir model best described adsorption of paracetamol drug on modified sugarcane bagasse with adsorption capacities ranging from 1.604 to 3.096 mg/g (Khan et al. 2012).

NH_4Cl-modified activated carbon derived from pomegranate fruit peels has been reportedly used to remove paracetamol from contaminated water at varied experimental parameters of pH (2–9), temperature (10–40 °C), and initial concentration (100–500 mg/L) (Mashayekh-Salehi and Moussavi 2016). Langmuir isotherm with $R^2 = 0.999$ gave the best description with a sorption capacity of 233 mg/g and removal percentage of 99.4%. The adsorption kinetics was well interpreted using pseudo-second-order kinetic model with a R^2 of 0.9981. Thermodynamic study showed that the adsorption process was feasible, exothermic, and spontaneous with a low degree of entropy (Mashayekh-Salehi and Moussavi 2016).

Activated carbon produced from thermally treated and microwave-activated tea waste has been utilized in removing paracetamol at varied operating conditions such as temperature (283–323 K), contact time (0–120 min), dose of adsorbent (1–2 g), initial paracetamol concentration (20–100 mg/L) and pH (3–8) (Dutta et al. 2015). At optimum conditions of pH (3), initial adsorbent concentration (100 mg/L), dose of adsorbent (1 g) and temperature (303 K), the maximum adsorption capacity of 195.95 mg/g and percentage removal of 99.42% were obtained (Dutta et al. 2015). Furthermore, adsorption isotherm and kinetics for removing paracetamol were excellently described with by Langmuir and pseudo-second-order kinetic models with respective R^2 of 0.993 and 0.999. Thermodynamic study showed that at a temperature range of 283 to 323 K, the adsorptive removal of paracetamol was endothermic and non-spontaneous with a high degree of entropy. In addition,

the desorption studies revealed a 97.97% recovery of adsorbent in a basic medium (Dutta et al. 2015).

H_3PO_4 has been used to modify Ayous sawdust and Cucurbitaceae peelings for removing phenacetin from simulated pharmaceutical wastewater at operating experimental parameters of 0.5 g of adsorbent and pH 2 (Ngakou et al. 2019). The obtained results showed the Langmuir maximum adsorption capacities of 15.43 and 5.33 mg/g for the Cucurbitaceae peelings and Ayous sawdust, respectively, with R^2 of 0.931 and 0.990 receptively. The adsorption isotherm was best described by Langmuir isotherm for the modified Cucurbitaceae peelings while Freundlich model best described the experimental data when ayous sawdust was used (Ngakou et al. 2019). In addition, both adsorbents were best described by pseudo-second-order kinetic model with respective R^2 of 0.852 and 0.943. Adsorption mechanism showed displayed a $\pi–\pi$ interactions of the phenacetin on modified ayous sawdust and Cucurbitaceae peelings (Ngakou et al. 2019).

Antibiotic drugs

Biomass-derived adsorbent prepared from rice straw has been used to remove ciprofloxacin and doxycycline at different operating conditions. The maximum adsorption capacity of 432.90 and 131.58 mg/g for doxycycline and ciprofloxacin, respectively, was obtained. Freundlich isotherm gave the best description of the adsorption with R^2 of 0.994 and 0.984, respectively. In addition, the adsorption kinetics was best fitted into the pseudo-second-order model with R^2 of 0.997 and 0.998, respectively, for doxycycline and ciprofloxacin, while the adsorption process was suggested to proceed through hydrogen bonding and $\pi–\pi$ interaction mechanism (Zeng et al. 2018).

Similarly, aqueous solution of KOH-modified pomegranate fruit peel has employed in removing ciprofloxacin at various adsorption conditions that include pH (2–12), contact time (15–150 min), dose of adsorbent (0.025–0.2 g) and initial concentration (50–300 mg/L). The experimental findings showed 86.4% removal efficiency and maximum sorption capacity of 2.353 mg/g. The Freundlich isotherm adsorption model gave the best description for the removal ciprofloxacin using KOH-modified pomegranate fruit peels with R^2 of 0.991 (Elhag Elhussien 2017).

Furthermore, biomass-derived char prepared from tea leaves at a temperature of 450 °C has been reportedly utilized to remove ciprofloxacin from aqueous solution at varied adsorption conditions of initial concentration (150–500 mg/L), pH (4–10), temperature (30–60 °C) at different time interval for 24 h (Li et al. 2018b). The sorption model that gave the best fit was Langmuir isotherm model and it showed a maximum adsorption capacity of 238.10 mg/g at optimum adsorption conditions, while the

sorption kinetics was fitted into the pseudo-second-order kinetic model. The adsorption process was feasible and controlled by hydrogen bonding, $\pi–\pi$ and electrostatic interaction mechanisms (Li et al. 2018b).

Thermal activation of Chinese herbal medicine (*Astragalus mongholicus*) waste at a temperature of 800 °C to prepare biomass-derived adsorbent for remediation of ciprofloxacin contaminated water has been reported (Shang et al. 2016). Under the optimum condition of pH at 7.0, the sorption capacity of 43.668 mg/g was obtained from the Langmuir model with R^2 of 0.999. Furthermore, pseudo-second-order model with R^2 of 0.999 gave the best interpretation of the adsorption kinetics. The mechanistic studies on the adsorption removal of ciprofloxacin onto the thermally activated *Astragalus mongholicus* residue was observed to be controlled by hydrogen bond, hydrophobic, electrostatic and $\pi–\pi$ interactions (Shang et al. 2016).

Batch adsorption studies at various operating conditions that comprises of initial concentration (0.5–70 mg/L), adsorption contact time (5–60 min), pH (2.5–9.5) and 0.1 g adsorbent dose has been investigated on removing ciprofloxacin from aqueous solution by using bamboo charcoal prepared by burning bamboo wood at a temperature of 800 °C in a kiln (Wang et al. 2017). At optimum pH of 5.5, bamboo charcoal exhibited maximum adsorption capacity of 36.02 mg/g for removing ciprofloxacin from an aqueous solution. Langmuir isotherm best described the adsorption with R^2 of 0.97. The adsorption kinetics for removing ciprofloxacin using bamboo charcoal was best interpreted using pseudo-second-order kinetic model. Furthermore, adsorption mechanism involving the ion exchange and hydrogen bonding was controlled the adsorptive removal of ciprofloxacin on bamboo charcoal. In addition, the presence of metal ion was also observed to influence the adsorptive removal of ciprofloxacin from aqueous solution (Wang et al. 2017).

The modification of Indian almond leaf has been reported for removing dicloxacillin from pharmaceutical aqueous solution. It was suggested that Langmuir isotherm model provided the best description of the adsorption of dicloxacillin on the modified Indian almond leaf with a high R^2 of 0.9650. A maximum adsorption capacity of 71.04 mg/g with a percentage removal of 86.93% was attained at optimum experimental adsorption parameters of pH (6.0), adsorbent dose (0.1 g), initial concentration of dicloxacillin (20 mg/L), temperature (283.15 K) and contact time (24 h) (Sunsandee et al. 2020). Additionally, the adsorption kinetics was suggested to be favoured by pseudo-second-order kinetic model with a R^2 of 0.9983 when compared to the pseudo-first-order kinetic model. Thermodynamic studies revealed that the adsorption process was feasible, spontaneous and exothermic with a poor degree of entropy proceeding with a mechanism of intermolecular

attraction force such as van der Waal force and hydrogen bonding (Sunsandee et al. 2020).

The modification of sugarcane bagasse with H_2SO_4, NaOH and urea has been reported as an economical adsorbent for removing levofloxacin from aqueous solution at various adsorptive experimental operating conditions (Khan et al. 2012). They reported that adsorptive removal of the drugs increased proportionally as the initial drug concentration increased from 2 to 8 ppm, beyond which there was no positive change in the adsorption rate of levofloxacin. Similarly, when 8 mg/L was used as the optimum drug concentration at varied adsorbent dose, a linear response relationship between the quantity of levofloxacin adsorbed and the dose of the modified sugarcane bagasse was noticed. This suggests that the adsorption capacity of the adsorption process increases with an increase in the adsorbent dose (Khan et al. 2012). The adsorption of the levofloxacin from aqueous solution attained equilibrium time within 180 min. It was opined that increase in levofloxacin adsorption with contact time was as a consequence of the availability of active site on the adsorbents. While a decrease in the amount of levofloxacin removed beyond the optimal contact time resulted from the rate of exhaustion of the active sites on the activated carbon. The Langmuir model was described to deliver the best fit for understanding the adsorption of the levofloxacin on the modified sugarcane bagasse with adsorption capacities in the range of 0.847–2.0465 mg/g for levofloxacin antibiotic drug (Khan et al. 2012).

Non-steroidal anti-inflammatory drugs

Bean (*Phaselous vulgaris*) husks which are usually disposed as debris that forms heaps are of environmental concern has been regarded to be edible to livestock especially in its fresh form but not edible to humans. These wastes are commonly available and are said to be economically viable materials for preparing the adsorbents. In recent times, there has been a quest for the production of activated carbon from biomass and this was the basis for preparing activated carbon from bean husk as an alternative for expensive commercial activated carbon (Bello et al. 2019c). The application of bean husk modified with ortho-phosphoric acid has been documented as an auspicious adsorbent for removing ibuprofen from water has been reported. It was estimated that at a pH of 4.75 and temperature of 50 °C, a maximum adsorption capacity of 50.0 mg/g was recorded using functionalized bean husk for removing ibuprofen from aqueous system. Langmuir adsorption isotherm gave best interpretation, while pseudo-second-order explained the adsorption kinetics model, while thermodynamic studies showed that the process proceeded through an endothermic and spontaneous route. Moreover, a desorption study using HCl, H_2O and NaOH revealed that the ortho-phosphoric acid functionalized bean husk exhibited a higher degree of regeneration and re-usability for removing ibuprofen from wastewater (Bello et al. 2019c).

The employment of ortho-phosphoric acid to modify orange peels and application in removing ibuprofen under different adsorption parameters showed a Langmuir maximum adsorption capacity of 49.30 mg/g at a temperature of 50 °C. Also, thermodynamics confirmed that adsorption process was feasible, spontaneous, and exothermic in chemical nature. Furthermore, kinetic studies revealed that adsorption process occurred via pseudo-second order with R^2 of 0.999. The process was suggested to occur by physisorption mechanism (Bello et al. 2020b).

Antimalaria drugs

Banana has been considered as the second largest quantity of produced fruits from the total world's production of fruits. Banana peels have been recognized to be a food waste that was classified as non-recyclable with a 1:2 ratio of banana product to waste. However, there is much more to do with the non-recyclable banana peels, and this includes composting, extraction of valuable organic compounds from banana peel in addition to preparation of activated carbons for several applications such as in wastewater treatment. For instance, the utilization of ortho-phosphoric acid-modified banana stalk for the removal of pharmaceutical contaminants such as lumefantrine drug from aqueous solution at varied operating parameters that include pH (3–11), contact time (0–200 min), initial lumefantrine concentration (20–100 mg/L), temperature (303–323 K) and 0.1 g adsorbent dose has been investigated (Agboola et al. 2021). The experimental results revealed that at optimum pH of 6, contact time of 120 min and temperature of 303 K, the maximum Langmuir adsorption capacity of 102.1 mg/g with R^2 of 0.9998 was achieved. Furthermore, pseudo-second-order kinetic model with R^2 ranging from 0.9845 to 0.9997 provided the best interpretation for the adsorptive removal of lumefantrine on chemically modified banana stalk. Additionally, thermodynamic studies showed that the process was spontaneous, feasible, and endothermic proceeding through intraparticle diffusion and boundary layer effect mechanisms (Agboola et al. 2021).

Modified biomass-derived adsorbents for dyes removal

Azo dyes

The quest for a cheap source of activated carbon as substitute to expensive commercial activated carbon in the treatment and remediation of aquatic ecosystem from environmental pollution with dyes has resulted in the employment of plant biomass as starting material for the synthesis of economically friendly activated carbon. For

example, modified coconut mesocarp has been utilized for removing organic pollutants such as azo-anionic dyes of Congo red and tartrazine from simulated wastewater. Effects of various experimental factors including initial concentration (40–100 mg/L), adsorbent dose (5–12 mg/L), etc. were investigated (Cocos 2021). It has been reported that modified cationic cellulose derived from coconut mesocarp yielded adsorption capacities of 19.61 mg/g and 19.99 mg/g for removing tartrazine and Congo red from simulated wastewater. Freundlich and Dubinin–Radushkevich models were, respectively, employed to identify adsorption isotherm process of tartrazine and Congo red. In addition, pseudo-second-order model gave a worthy depiction for the adsorption kinetics in tartrazin and Congo red with R^2 of 0.999 for the adsorptive removal from wastewater (Cocos 2021).

The modification of lemon grass to produce viable adsorbent for removing methyl red has been investigated. The experimental outcome revealed that modification of lemon grass with ortho-phosphoric acid yielded activated carbon with desirable properties such as 836.4 m^2/g surface area, 598.06 m^2/g mesopore surface area, with a total pore volume and average pore diameter of 0.472 cm^3 and 3.62 nm, respectively (Ahmad et al. 2019). Effects of various operating parameters such as 25–500 mg/L initial concentration, 0–24 h contact time, 30–60 °C temperature and 2–12 solution pH revealed that the removal of methyl red using lemon grass was spontaneous and endothermic. It proceeded via physisorption process with 63.87% percentage adsorptive removal and 76.923 mg/g maximum monolayer adsorption capacity as described using Langmuir isotherm with R^2 of 0.983, while kinetic studies confirmed the favourability of pseudo-first-order kinetic model (Ahmad et al. 2019).

Removal of methylene blue has been successfully experimented using H_2SO_4-modified coconut shell. It was estimated that optimum adsorption capacity of 50.6 mg/g was achieved when H_2SO_4-modified coconut shell was utilized as tremendous adsorbent for removing methylene blue from wastewater. The adsorption kinetics and isotherm were best described using pseudo-second-order and Freundlich models, respectively. The mechanism of adsorption process occurred via hydrogen bonding, π–π, and electrostatic interactions (Jawad et al. 2020).

Acid-activated *Posidonia oceanica* has been reportedly utilized for the remediation of methylene blue from spiked brackish wastewater from a lake in Egypt called Manzala (Elmorsi et al. 2019). From the batch adsorption findings, it was suggested that at optimum experimental operating conditions, there was an adsorption percentage removal in the range of 91.5–99.9%. Furthermore, when compared with the Freundlich and Langmuir isotherm models, the experimental findings suggested that the

Dubinin–Radushkevich model gave the best description for the adsorption isotherm with a R^2 of 0.992 (Elmorsi et al. 2019).

Acid/thermal-modified rice husk has also been used for removing methylene blue with a percentage removal of 96.7% and maximum Langmuir adsorption capacity of 103.11 mg/g with R^2 of 0.9962 (Moeinian and Mehdinia 2019). Similarly, the use of modified lemon grass has been employed in removing methylene blue from aqueous environment (Ahmad et al. 2021a). It was reported that maximum percentage removal of 64.34% methylene blue was recorded at optimum pH 12 and maximum sorption capacity of 342.9 mg/g. Koble-Corrigan model isotherm best described the adsorption with a R^2 of 0.999. The adsorption process was endothermic and its adsorption kinetics was best described by pseudo-first-order kinetic model proceeding via physisorption mechanism (Ahmad et al. 2021a).

KOH-modified pomegranate fruit peels have been used in removing methylene blue dye from simulated wastewater under the influence of operational conditions, including temperature (303–313 K), initial methylene blue dye concentration (25–300 mg/L) and contact time (0–24 h) (Ahmad et al. 2021c). The percentage removal of 83.4% removal and a maximum Langmuir sorption capacity of 235.58 mg/g with a R^2 of 0.977 were obtained. Freundlich isotherm model gave the best interpretation of the isotherm adsorption model with a R^2 of 0.998 (Ahmad et al. 2021c). Compared to pseudo-second order, the sorption kinetics was well interpreted using the pseudo-first order at a temperature range of 303–312 K. The sorption process was observed to be feasible, exothermic, and spontaneous proceeding through a physisorption mechanism of adsorption (Ahmad et al. 2021c).

The utilization of polyvinyl alcohol (PVA) coated activated carbon derived from the stems of *Crotolaria burhia* and *Opuntia dellinii* as adsorbents have been reported in removing azo dyes that include methylene red and methylene blue from aqueous solution (Gehlot et al. 2009, 2011). It was noted that the PVA coated activated carbon displayed 89.1% removal efficiency and a maximum Langmuir adsorption capacity of 19.23 mg/g with a R^2 of 0.9975. The adsorptive removal of azo dyes using PVA coated activated carbons derived from the stems of *Crotolaria burhia* and *Opuntia dellinii* was excellently described by Freundlich isotherm model with a R^2 of 0.994 (Gehlot et al. 2009, 2011).

The modification of wheat husk with 30% hydrogen peroxide for 24 h, which was preceded by washing with distilled water then oven drying at 60 °C, has been investigated in removing Reactofix Red 3BFN from aqueous solution. The investigated batch adsorption conditions are adsorbent dose (4.0–24.0 g), pH (2–10), initial dye concentration (1.0×10^{-5}–6.0×10^{-5} mg/L) and temperature (30–50 °C) (Jain et al. 2006a). The results obtained from the

experimental findings showed that at optimum pH of 2.0, the modified wheat husk displayed 90% removal of Reactofix Red 3BFN. At an optimum temperature of 30 °C, there was a maximum adsorption capacity of 80.37 mg/g with a R^2 of 0.962. Furthermore, thermodynamic studies showed that at temperature range of 30–50 °C, the batch adsorptive removal of Reactofix Red 3BFN from an aqueous solution using H_2O_2-modified wheat husk followed a spontaneous, feasible, and endothermic chemical path (Jain et al. 2006a).

Xanthene dyes

The utilization of ortho-phosphoric acid functionalized coconut husk in sequestering Rhodamine B dye has been demonstrated to achieve a maximum Langmuir adsorption capacity of 1666.67 mg/g with R^2 of 0.99. Furthermore, premised on the thermodynamic and kinetic studies, the adsorption process was said to be chemically endothermic and spontaneous proceeding via a pseudo-second-order adsorption kinetic model (Bello et al. 2019a).

Similarly, the adsorptive removal of Rhodamine B dye using functionalized locust bean pod has been investigated at varied experimental conditions of Rhodamine B dye concentrations (200–1000 mg/L), dose (0.1 g), temperature (303, 313, 323 K), and agitation speed of 120 rpm. The adsorption mechanism of using ortho-phosphoric acid-modified locust ban pod for removing Rhodamine B dye proceeded via physisorption mechanism with endothermic, spontaneous, and feasible reaction path. This yielded maximum monolayer adsorptive removal capacity of 1111.1 mg/g as deduced from Langmuir isotherm that best interpreted the adsorption. The adsorption kinetics was best described with pseudo-second-order kinetic model (Bello et al. 2019b).

In addition, the use of functionalized *Gmelina aborea* leaf for removing Rhodamine B dye from water at experimental conditions of pH, temperature, adsorbent dose, initial dye concentration and contact time has been reported to yield maximum adsorption capacity of 1000 mg/g (Bello et al. 2020a). Additionally, adsorption kinetics was best described with pseudo-second-order kinetic model and thermodynamics study revealed that sequestering of Rhodamine B dye using modified *Gmelina aborea* leaf was characterized by an endothermic, spontaneous, and feasible chemical process that proceed via a physisorption mechanism. Moreover, from the recycling and regeneration studies using HCl, H_2O and NaOH revealed that 0.5 M HCl for desorbing Rhodamine B dye from modified *Gmelina aborea* leaf yielded a regeneration efficacy that increased from 8.26 to 92.74% (Bello et al. 2020a).

The employment of modified mango pod has been investigated in removing Rhodamine B dye from aqueous media at varied operating conditions which include pH (2–9), adsorption contact time (0–120 min), dose of adsorbent (0.1 g), temperature (303–323 K) and initial Rhodamine B dye concentration (200–1000 mg/L) (Bello et al. 2021). Adsorption kinetics and isotherm were best described via pseudo-second-order kinetic model and Freundlich isotherm model, with R^2 of 0.99 (Bello et al. 2021). Maximum adsorptive removal capacity of 500 mg/g was obtained and thermodynamic studies showed that the adsorptive removal process was feasible, endothermic, and spontaneous proceeding under the influence of both intraparticle diffusion and boundary layer mechanisms. In addition, the cost analysis revealed that the utilization of modified mango pod to prepared adsorbent with a cost price of 34.20 USD/kg in removing Rhodamine B dye from aqueous media was less expensive compared to commercial activated carbon with a cost price of 259.5 USD/kg, thereby translating to about 8 times less expensive compared than the commercial activated carbon (Bello et al. 2021).

Miscellaneous diagnostic dyes

The modification of rice husk by treatment with hydrogen peroxide preceding the acetic acid treatment to prepare a vital adsorbent has been investigated in removing indigo carmine dye from aqueous solution at various conditions of pH (2–12), initial dye concentration (1.0×10^{-4} –9.0×10^{-4} mg/L), temperature (40–60 °C), dose of adsorbent (0.5–3.5 g) and contact time (10–70 min) (Jain et al. 2006b). Adsorptive removal of indigo carmine dye increased with increase in adsorbent dosage in addition to contact time, nonetheless, decreases with increasing initial dye concentration, solution pH and temperature. At an optimum temperature of 40 °C, a maximum adsorption capacity of 9.275 mg/g was recorded as obtained from Langmuir model. Pseudo-first-order model gave the best description for the adsorption kinetics at room temperature, fixed pH and adsorbent dose, while the thermodynamic studies showed that adsorptive removal of indigo carmine dye onto the modified rice husk was exothermic and spontaneous (Jain et al. 2006b).

Tri-aryl methane dyes

The modification of *Azolla filiculoides* with 0.1 M HCl has been studied as a feasible adsorbent for removing Acid Green 3 dye. The experimental findings demonstrated that the optimum conditions were contact time (90 min), pH (3), dose of adsorbent (4 g/L) and initial concentration (10 mg/L) (Balarak et al. 2016). The utilization of HCl-modified *Azolla filiculoides* exhibited 99.1% removal for Acid green 3 dye at optimum adsorption parameters. Moreover, the Langmuir adsorption isotherm model gave the best description of the adsorption process with maximum adsorption capacity of 37.5 mg/g and R^2 of 0.999. Adsorption kinetic proceeded

via pseudo-second-order kinetic model which displayed a R^2 of 0.999 (Balarak et al. 2016).

Phenol-derived polymeric dyes

The employment of thermally and chemically modified rice husk for the removal of phenol (a precursor for phenol-derived polymeric dyes) has been reported. The thermally treated rice husk exhibited higher surface of 24–201 m^2/g over the calcium hydroxide-treated rice husk with surface area of 3.19 m^2/g, and also displayed a higher adsorptive removal capacity of 36–64%, compared to 28% displayed by the calcium hydroxide-treated rice husk (Daffalla et al. 2020). The emergence of research on the use of modified biomass wastes has led to numerous studies in the recent years for their employment in wastewater remediation containing different organic pollutants as presented in Table 2.

Mechanistic aspects of organic pollutants adsorption

During adsorption process, the interactions between the surfaces of organic pollutants (adsorbates) and adsorbent proceeds until dynamic equilibrium is attained. There are three major processes that occur during the adsorption of an adsorbent by an adsorbate and they are as follows: physical adsorption, precipitation and complexation, and pore filling (Fagbohungbe et al. 2017). Physical adsorption is the stage at which adsorbates settle on the surface of adsorbent but any adsorption does not take place; hence, it is called the clear zone. On the other hand, the precipitation and complexation stage is caused by the deposition of the adsorbates on the surface coverage area of the adsorbent. This stage involves the mass transfer of adsorbates into the adsorbent with the adsorption progressing by chemical bonding. Therefore, it is called as the mass transfer zone. Pore filling stage is premised on the condensation and retention of adsorbate into pores of the adsorbent until equilibrium is attained, therefore it is referred the exhausted zone (Fagbohungbe et al. 2017; Ambaye et al. 2021). From these three stages can be observed the three critical zones which are as follows: clear zone, mass transfer zone, and the final stage which is referred to as exhausted/saturated zone, where equilibrium is attained (Ambaye et al. 2021). Inverse proportionality is observed between saturated zone and clear zone. Meanwhile, mass transfer zone remains unaltered, apart from the increase in adsorbate's concentration. This process continues until the adsorbent is saturated, and this point is called breakthrough point (Moreno-Castilla 2004). A removal mechanism for adsorption of organic pollutants onto biomass is presented in Fig. 3. Hydroxyl, carboxyl, carbonyl,
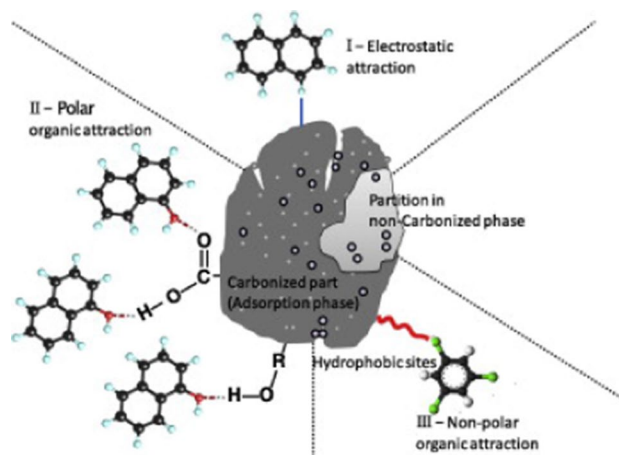

Fig. 3 Different adsorption mechanisms for organic contaminants. Partition/adsorption are indicated with circles on adsorbent's particles. I—electrostatic interaction between the modified biomass adsorbent and organic contaminant, II—electrostatic attraction between the modified biomass adsorbent and organic contaminant that are polar, and III—electrostatic attraction between the modified biomass adsorbent and non-polar organic contaminant (Ahmad et al. 2014a)

and amine, which are organic functional groups, favour the adsorption of organic molecules on the surface of the biomass. This is an example of a donor/acceptor electron type of adsorption mechanism based on the unbalanced electron distribution between the functional groups of the biomass and the organic compound. Notably is the highly reduced bonding between the adsorbent and organic compounds with active chloro- and nitro- substituent groups. Therefore, the binding energy between the biomass and the organic compound is increased (Mu'azu et al. 2017; Ambaye et al. 2021) due to the substituent group being a strong electron acceptor (Atkinson et al. 2010).

The mechanisms of adsorption of organic pollutants have been classified as hydrophobic interaction, pore filling, electrostatic interaction, partitioning, and electron donor–acceptor (EDA) interaction. The partitioning mechanism involves the diffusion of the adsorbates into the pores of the non-carbonized portion of the adsorbent due to their easy interaction between the organic adsorbates. Therefore, the adsorptive removal of the organic pollutants is reliant on the properties of the non-carbonized portion of the adsorbent, and the partitioning mechanism is highly effective when the adsorbent possesses high content of volatile matter in the presence of high concentration of organic pollutants (Ambaye et al. 2021). The electrostatic interaction mechanism is the most significant mechanism that takes place in the adsorption of ionizable organic compounds to adsorbents that exhibit positively charged surfaces through electrostatic interaction. However, the ionic strength as well as the solution's pH determines the

efficiency of the electrostatic interaction mechanism in attracting and repelling organic pollutants. This is attributed to the increase in repulsive electrostatic interaction between adsorbate and adsorbent when they both possess same charges (Ambaye et al. 2021).

The pore filling mechanism occurs by the passage of the organic pollutants through the micropores and mesopores of the adsorbent but the mechanism is dependent on the polarity of the organic pollutants as well as the nature and type of the adsorbent. However, the efficiency of the pore filling mechanism depends on the occurrence of petty content of volatile matter in the presence of low concentration of organic pollutants. The electron donor and acceptor interaction mechanism frequently occurs in the adsorptive interaction of aromatic compounds on adsorbents that exhibit a graphene-like structure, which is only attained at a temperature above 1100 °C (Ambaye et al. 2021). Nevertheless, the pyrolysis temperature employed in the fabrication of the adsorbent determines the development of enriched or deficient π–electron adsorbent. That is, system of adsorbent's π aromatic compound acts as electron acceptor and donor when the adsorbents are obtained at temperature above and below 500 °C. The hydrophobic interaction is habitually employed in adsorptive removal of neutral and hydrophobic organic compounds through the portioning as well as hydrophobic interaction processes, but requires less amount of energy in comparison with the partitioning mechanistic process. This is subsequent to the decline in the quantity of polar functional groups on the surface of the adsorbent as an outcome of pyrolysis temperature (Ambaye et al. 2021).

Regeneration studies and environmental challenges with chemical modifications

The regeneration of adsorbent used is dependent on the source of the adsorbent, regeneration method, and adsorbent dosage. During a regeneration study, the recovery of the adsorbates can be achieved through the principles of desorption and decomposition (Jia et al. 2013; Wang et al. 2015; Ambaye et al. 2021; Bello et al. 2021). The regeneration studies on the use of eucalyptus leaves to prepare magnetic biomass for decontamination of organic pollutants revealed that the FTIR spectra band shift did not change until six continuous cycles (Wang et al. 2015). They attributed this observation to the increment in the ash content of the prepared biomass as well as the effect of surface functional groups of the biomass that did not change significantly. The pore sizes and surface area of the prepared and regenerated magnetic biomass are noticed to be similar, thus allowing high separation and adsorptive rates. The process of adsorption is made more economical by the regeneration method. It was further observed that the

regeneration study using a basic chemical such as NaOH is more efficient. Another method of chemical regeneration is altering the pH of adsorbents to desorb substances that are non-reactive, like dyes and aniline (Fagbohungbe et al. 2017). However, major challenges associated with regeneration studies using chemical methods are the high cost of some required reagents and the potential to aggravate environmental pollution. Based on this, the disposal of digestate is therefore very important in the management of biomass.

Recently, thermal regeneration method has been recognized as the best technique for the recovery of biomass since it permits the development of tiny pore sizes contrasted with the first pores of the initial biomass, mainly when it is completed at high temperatures. The researchers noticed that temperature increase improved regeneration efficiency. For instance, in a study on removal of organic compounds such as pyrene and benzopyrene using biomass adsorbent derived from *Enteromorpha prolifera* and the regeneration studies carried out at varied temperatures of 80 °C, 150 °C, and 200 °C, an increased in temperature improved the regeneration efficiency (Qiao et al. 2018). The adsorption efficiency of pyrene was observed to increase by 35.0%, 45.0%, and 48.0%, while that of benzopyrene increased by 31.0%, 41.0%, and 40.0%, respectively (Qiao et al. 2018). Furthermore, it was also observed that dissolved organic carbon could be reduced by biomass regeneration using the thermal method.

Regeneration of biomass had also been carried out using irradiation by microwave method. Results obtained show that the biomass regeneration was very rapid. Additionally, it allowed easy control of the operating temperature. Inducement of polar molecules in the biomass was observed when it was irradiated by microwave, thus leading to the formation of dipole/polarization. However, this method is still at the experimental stage and laboratory scale because its application on an industrial/large scale is currently absent (Jia et al. 2013; Ahmad et al. 2014b, 2021b; Khasri et al. 2018; Li et al. 2018a; Ambaye et al. 2021; Yusop et al. 2022). Recently, the operating pressure was adjusted using supercritical fluid in order to investigate biomass regeneration (Hu 2018; Dai et al. 2019). It was observed by the authors that no change occurred in the physicochemical properties of the biomass when used to adsorb high molecular weight volatile organic compounds. This method shows advantages such as minimal loss of biomass, a short operating cycle, and low operating temperatures. However, because of the complexity of the method, the costly materials, equipment, and high-pressure resistance, the method is still at the experimental stage and needs to be scaled up. In addition, of all the methods used for the regeneration and management of biomass in the previous studies, the thermal regeneration process seems to

be most efficient due to minimal operation cost and maximal economic applicability. However, carbon loss is very high when compared with other methods. Further research is required which should focus on the economic value of biomass used for adsorption.

Realizing that the use of chemical modification has become very popular for improving the adsorption uptake of organic pollutants, several studies conducted for adsorption of various organic pollutants confirmed that this approach is feasible for translating the applicability of modified biomass adsorbents into industrial application. This is based on the efficient adsorbent properties such as availability of the biomass material at low/zero cost, enhanced porosities, and larger surface area. However, evaluating the effects that these modified adsorbents pose to ensure the global environmental safety is highly important. After a successful adsorption process, the life cycle experiment through desorption studies is necessary to ensure the number of cycles that these adsorbents are able to complete before ripping off of their maximum adsorption capacities. For waste disposal, different strategies such as catalytic technology, biological treatment, ion exchange and advanced bio-physical methods have been developed to eradicate the different imbalances that the spent (used) adsorbent pose to the immediate environment (Xu et al. 2017; Ogunlalu et al. 2021). The adsorbents that are not capable of been reused should be burnt off at a very higher temperatures such as 1100 °C to 1200 °C and the resultant gases should be collected using a specialized equipment (Zhou et al. 2008; Ogunlalu et al. 2021).

Computational (optimization) models for modified biomass adsorbents for removal of organic pollutants

Response surface methodology

The response surface methodology (RSM) has two most commonly used designs which are central composite design (CCD) and Box–Behnken design (BBD). The systematic utilization of RSM for the adsorptive removal of malachite green, methylene blue, azo and anthraquinone dyes from effluents has been reported (Song et al. 2018; Chowdhury et al. 2019; Ahmad et al. 2020). In addition, adsorptive removal of acid dye using derived activated carbon for antibiotic (ciprofloxacin) removal from wastewater using rice husk as well as other research findings on the employment of RSM for the adsorptive removal of organic contaminants from aqueous system has established the effective application of RSM as an efficient computational model in wastewater treatment (Amini et al. 2008; Niad et al. 2014; Samuel et al. 2015; Zhou et al. 2019).

RSM has been confirmed to be suitable in studying the removal of methyl orange using modified sugar beet bagasse and results obtained show concordance between experimental and predicted values. The optimum adsorption process conditions generated using RSM for the aqueous removal of methyl orange were 2.51, 100 mg/L, and 0.37 g/L for pH, initial methyl orange concentration, and adsorbent dose, respectively, with predicted adsorption capacity and adsorptive removal efficiency of 221.5 mg/g and 51.8%, respectively (Ghorbani and Kamari 2017). RSM using two-factor interaction models and quadratic models have been applied in optimizing the preparation of functionalized pomegranate fruit peels. Its application in removing Remazol brilliant blue R dye resulted in a 31.2% yield of activated carbon derived from functionalized pomegranate fruit peels and 81.35% removal of Remazol brilliant blue R dye. The model suitability was performed by means of analysis of variance. It was suggested from the percentage error between the actual and predicted result calculated to be 1.92% that there was a good correlation between experimental and predicted results (Ahmad et al. 2020).

In a similar study, RSM was used to model the adsorption of methylene blue (MB) from aqueous system using pomegranate fruit peel activated carbon (PFPAC) which was prepared by KOH impregnation and CO_2 gasification methods. Results obtained reveal that the optimum sorption conditions for the removal of MB dye are 375 W, 4.5 min, and 0.9, for radiation power, activation time, and impregnation ratio, respectively. These optimum adsorption conditions resulted in 83.4% removal of MB dye and PFPAC yield of 30.8%, respectively (Ahmad et al. 2021c). Similarly, use of eggshell in the adsorptive removal of Malachite green (MG) dye was studied using CCD to investigate effects of operating conditions on the adsorptive removal of the dye. There was significant agreement between predicted values obtained with experimental results at R^2 value of 0.9388. The optimum adsorptive removal was 90.66% at operating parameters of 1.25 g adsorbent dosage, pH 6, and contact time of 40 min. Results from this study further revealed that the eggshell could be a good adsorbent for MG removal from aqueous solution (Hoo et al. 2022). The second-order polynomial equation of CCD has been reportedly utilized in the optimization of adsorption parameters for the adsorptive removal of methylene blue, brilliant green and Congo red with their variable levels. The predicted optimum adsorption condition was 30 °C, 9.8, 2.5 g/L and 99% for temperature, pH, adsorbent dose, and percentage adsorptive removal of dye using CCD. The qualitative fit of the model was defensible by the R^2 of 0.9966, which exceeded the R^2 of 0.88 suggested as best correlation from previous studies (Fegousse et al. 2019). Grape leaves prepared activated carbon was used for the removal of MB from aqueous solution. Data modelling and design of the experiment were

carried out using RSM. The results obtained show that the optimum percentage removal of MB was 97.4% at adsorption conditions of adsorbent dose 12.5 g/L, MB concentration of 100 mg/L, pH 11, and contact time of 90 min. Furthermore, obtained results indicate that adsorbent dosage and initial dye concentration play major role in increasing the efficiency of MB adsorption (Alireza et al. 2022).

Green pea which is a low-cost adsorbent was investigated for the adsorptive removal of Reactive Blue 19 dye using Box–Behnken design to deduce the effect of independent and interaction influences of process variables such as temperature, pH, and adsorbent dosage. The RSM experimental results revealed that maximum removal was 99.42% at optimum conditions of temperature (35 °C), pH 2, and adsorbent dosage (1.5 g/100 mL). Furthermore, the kinetic study showed that Freundlich isotherm fitted best for the removal of the dye and it followed pseudo-second-order model (Demirhan 2020).

The adsorptive removal of Direct Violet 51 dye from aqueous solution by biosorption using sugarcane bagasse biomass was conducted by different physical and chemical treatments (Sadaf et al. 2015). Box–Behnken experimental design was used to ascertain the effect of three independent variables (biosorbent dose, pH, and initial dye concentration). Optimum removal of Direct Violet 51 dye was 63.0 mg/g and was attained at 0.05 g biosorbent dose and pH 2. Desorption study on the loaded biosorbent using 1 M NaOH solution removed 61.58% dye (Sadaf et al. 2015). Additionally, the use of plackett–Burman design and RSM has been employed in the statistical optimization of *Gracilaria edulis* for adsorption removal of dye from textile effluents (Venkataraghavan et al. 2020).

Artificial intelligence

Owing to current technological advancements, there has been upsurge in the application of artificial intelligence (AI) model for various applications in different fields including water purification, catalysis, medical, etc. Interconnected assembly of units called artificial neurons or nodes is used for the design of AI and this concept is premised of biological nervous system. Currently, this concept enjoys global attention as a computational-based modelling tool which is highly efficient for predicting rate of pollutant removal using sets of data. This provides avenue for comparative study of the correlation coefficient obtained from experimental and expected data. This leads to a reduction in the cost and time of an experiment. Furthermore, AI has been shown to be capable of identifying complex and complicated nonlinear relationships among several parameters and variables in

addition to showing correlations of simulated output and the interactive influence among variables (Nasr et al. 2017; Mazloom et al. 2020; Liao and Yao 2021; Martini and Roni 2021).

For example, use of artificial neutral network (ANN) has been reported to model pseudo-second-order kinetic model for adsorptive removal of paracetamol using modified orange peels from aqueous media at different adsorption conditions such as initial concentration (10–50 mg/L), contact time (0–330 min) and temperature (30–50 °C) (Afolabi et al. 2020b). It was revealed that the ANN with hyperbolic tangent sigmoid transfer function at the input layer and linear transfer function at the output layer, eighteen (18) hidden neurons and Levenberg–Marquardt as its backpropagation algorithm showed the optimal prediction ability. It further demonstrates the effective application in modelling pseudo-second-order kinetic model using modified orange peels in removing paracetamol (Afolabi et al. 2020b). Several, other report on the application of AI for modelling the efficiency of organic adsorption process has been reported in the past few studies. Table 3 presents various adsorption performance comparison of different adsorbents using Artificial intelligent.

Concluding remarks

The purification of water and wastewater using adsorption technique is a globally recognized and reliable scientific procedure to overcome the predicament of contaminant with most traditional techniques applied in wastewater treatment operation. Adsorption has been ascertained to be economical, safe, and stress-free to employ. These characteristics have abetted it to prevail over the impediments that include enormous consumption of space, extravagant, commercially unappealing, and operational toil, in addition to the disposal glitches which is common with conventional techniques. The obtainability of enormous quantities of naturally existing materials in addition to wastes generated from both agricultural activities has gotten numerous considerations in contemporary time. The extensive employment of these potential adsorbents after subsequent modification by means of more than a few techniques that include chemical, mechanical, thermal, gasification, or blend of techniques.

Efficient biomass-derived adsorbent is expected to be well-designed and serves as good candidate for efficient removal of organic pollutants from wastewater treatment plant. Based on the survey of literature, use of unmodified biomass adsorbents for decontamination of organic pollut-ants from aqueous mixture is not certain and suffers poor

Table 3 Comparison of the various adsorbent performances using AI for the adsorptive removal of organic pollutants

Adsorbents	Pollutants	Optimum number of neurons	Evaluation indices						References
			R^2	MRE	AE	MSE	RMSE	SSE	
AC from *Acacia auriculiformis*	Direct blue 86	5-8-1	0.982						Dutta et al. (2012)
AC from coconut shell	Indigo dye	6-5-5-1					0.07283(train), 0.10699(test)		Deshmukh (2012)
AC from orange wood tree	Sunset yellow	3-13-1 3-15-1	0.997 0.996			0.0001 0.0001			Ghaedi et al. (2015)
Activated carbon	Methylene blue	–			0.0014				Xu and Hu (2010)
Activated spent tea	Methylene blue	5-10-1	0.999						Babaei et al. (2016)
Au NP AC Tamarisk	Methyl orange	3-11-1 3-20-1	0.958 0.989			0.00082 0.0006			Ghaedi et al. (2014)
Bamboo biochar	Acid black 172	5-5-1	0.996			1.05×10^{-4}			Yang et al. (2014)
Chara contraria	Lanaset Red G	4-23-1	0.999						Çelekli et al. (2012)
Eucalyptus camaldulensis)	Basic Blue 41 Reactive Black 5	3-5-1				0.006206 0.001193			Balci et al. (2011)
Iron-coated pumice	Ethidium bromide	4-1-2-1	0.999			0.005			Heibati et al. (2016)
Modified rice straw	Crystal violet	3-17-1	0.997						Chakraborty et al. (2013)
NaOH-modified rice husk	Methylene blue	3-13-1	0.995						Chowdhury and Saha (2013)
Orange peel	Paracetamol		0.99						Afolabi et al. (2020a)
Posidonia oceanica L	Methylene blue	3-15-1	0.998						Cavas et al. (2011)
Potato peel powder	Reactive Red 198	5-7-1	0.98				4.3		Maleki et al. (2013)
Powdered activated carbon	Acid orange 7	3-2-1		5.81%					Dutta et al. (2012)
Rice husk	Safranin onto	3-10-1	0.988						Saha et al. (2012)
Rice husk carbon	Bromocresol red Alizarin red Malachite green Methylene blue	3-5-5-2					0.025(train) 0.024 (test)		Khonde and Pandharipande (2012)
Sawdust	Dye and metal ion		0.99						Dolatabadi et al. (2018)
Soya bean waste	Methyl violet 2B	6-4-1	0.995						Kooh et al. (2016)
Walnut husk	Lanaset red G	4-20-1	0.995			0.499–5.006			Çelekli et al. (2012)
Walnut husk	Basic Red (BR) 46	5-25-1	0.999					0.5848	Çelekli et al. (2016)
Walnut husk	Azo dye		0.999			0.2303			Çelekli et al. (2016)
Waste marble dust	Malachite green Acid blue 161	4-12-1	> 0.89			< 0.01			Çoruh et al. (2014)

MAE Mean average error, *MSE* Mean square error, *AE* Average error, *RMSE* Root mean square error, R^2 Correlation coefficient, *MRE* Mean relative estimation error

surface area, pore volume and sizes while their adsorption performance is also low. However, it has been demonstrated by previous studies that several types of modifications seemed to enhance the biomass adsorbents for better adsorptive performance towards organic pollutants (including pesticides, insecticides. pharmaceutical and dyes). Furthermore, literature has shown that many of these studies based on the adsorption of organic contaminants using biomass adsorbents were carried out in aqueous systems and

on a laboratory scale. This did not undermine the potentiality of these studies. However, for commercial and practical applications, adsorbents from biomass origin should be effectively applied directly into effluents of wastewater. Therefore, more studies should be carried out to elucidate the design and synthesis of biomass-based adsorbents which can be used successfully in wastewater treatment systems for decontaminating organic pollutants.

Additionally, focus of future studies should be on sustainable measures on how spent biomass adsorbents that have lost their performance/ability on organic pollutants would be effectively disposed. This is to ensure proper understanding of the stability of biomass-based adsorbent materials. Also, it is necessary to corroborate the commercial values and the practical applications of modified biomass materials towards organic pollutants removal in wastewater and their vital associated issues in the environment.

Furthermore, in recent years, use of combined processes has received increased attention for decontaminating organic pollutants from aqueous mixtures. There is, however, insufficient knowledge and understanding on combined adsorption process and existing treatment technologies like reverse osmosis, nanofiltration, ozonation, Fenton-oxidation, and photocatalytic degradation. Hence, investigation on the efficiency of removing organic pollutants using hybridized adsorption process and other treatment methods is necessary. Future studies should be centred on designing and production costs of modified adsorbents from biomass source and the corresponding cost evaluation analysis for organic pollutants contaminant treatment from laboratory, pilot, and industrial scales. This should help to establish viable method(s) that should be implemented for disposing the spent adsorbents. Such technique is expected to be environmentally sustainable, and hybridized adsorption with other treatment processes (methods) to ensure highly and better removal performance. The limitations that prevent most biomass adsorbents from becoming commercially viable can be reduced by investigating these knowledge gaps.

In agreement with the green/sustainable chemistry, since chemical modification significantly enhances the sorption capacity, when compared with the unmodified counterpart, therefore use of environmentally friendly reagents should be focused on with aim of minimizing the use of these chemicals and minimization of the wastes generated and released into the environment.

The effects of interfering ions present in the environment laden with organic pollutants are not considered in many chemically modified biomass materials. Presence of interfering ions such as NO_3^-, Cl^-, SO_4^{2-}, $H_2PO_4^-$, Ca^{2+}, and Fe^{2+} have been previously reported to interference with the organic contaminants (pesticides including endosulphan, 2,4-dichlorophenoxyacetic acid, humic acid and atrazine) (Das et al. 2009). More investigation on the use of these sorbents for systems with the multiple organic pollutants in the same environments is required to examine the influence of one organic pollutant adsorption over the other. Similarly, further studies are needed on the chemical(s) that can introduce certain functional group(s) that are capable of adsorbing different contaminants same way they are present in the real wastewater system and the corresponding influence of one contaminant on the uptake of the others.

Lastly, the utilization of biological (biomass) materials as precursor for the fabrication of nanoparticles (NPs) is currently attracting wider popularity as a secondary metabolite owing to their inherent properties as reducing agents for the NPs and stabilizing/capping agents. Interestingly, most of the resultant NPs have reduced particle sizes, higher pore volumes and surface areas that were better than their precursors (Yu et al. 2016; Iwuozor et al. 2021). This suggests that more research in the area is promising to optimize the usage of chemical substances on the sorbents for sorption process.

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Data availability All data and materials used in this study are available within this article.

Declarations

Conflict of interest The authors declare that they have no conflict of interest.

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