Green Adsorbents for Environmental Remediation: Synthesis Methods, Ecotoxicity, and Reusability Prospects

Yanju Liu 1,2,*, Bhabananda Biswas 1,2, Masud Hassan 3 and Ravi Naidu 1,2

1 Global Centre for Environmental Remediation, ATC Building, Callaghan Campus, Newcastle, NSW 2308, Australia; bhaba.biswas@newcastle.edu.au (B.B.); ravi.naidu@newcastle.edu.au (R.N.)
2 crcCARE Pty Ltd., ATC Building, Callaghan Campus, Newcastle, NSW 2308, Australia
3 College of Resources and Environmental Engineering, Guizhou University, Guiyang 550025, China; masud.hassan@gzu.edu.cn

* Correspondence: yanju.liu@newcastle.edu.au

Abstract: Adsorbent materials have long been used for remediating environmental contaminants. There is an increasing focus on developing sustainable adsorbent materials for long-term use in environmentally friendly and cost-effective remediation. “Green” or “eco-friendly” sorbent materials are generally prepared from renewable or recycled resources, have minimal toxic effects, involve synthesis processes with minor chemical or energy footprints, have high reusability, and do not contribute to additional waste or contamination. Thus, it is essential for materials to have high sorption capacity, high stability, and reusability. The literature focuses on using low-cost or waste materials to produce sorbent materials for the immobilization of contaminants from soil and water systems. The regeneration possibilities of adsorbents are used to evaluate their cost effectiveness and long-term environmental impact once they are applied at field-scale. This review evaluates sustainable sorbent materials, highlighting their green and eco-friendly qualities for a circular economy, and their contribution to the United Nations Sustainable Development Goals (UNSDG). The synthesis techniques, ecotoxicity, and prospect of reusing adsorbents are highlighted. Further, the review provides insights for researchers and practitioners interested in developing and applying green adsorbents, including bio-based carbon, char, and fibrous materials for soil and water remediation.

Keywords: green synthesis; eco-friendly adsorbent; adsorption; degradation; environmental remediation; reusability

1. Introduction

Exposure to environmental pollutants from soil and water poses potential risks to biotic and abiotic ecosystems [1–4]. These pollutants are mainly manufactured chemicals, including heavy metals, fluoroalkyl compounds, agrochemicals, pharmaceuticals, and numerous known and unknown chemicals [5–7]. Remediating these pollutants is the priority for ensuring a sustainable ecosystem, and adsorption has become a promising technology due to its efficiency, biocompatibility, and low operating costs [5,6,8–11]. For example, adsorption has been proven to be effective for treating soil and wastewater containing toxic heavy metals, dyes, antibiotics, per- and polyfluoroalkyl substances (PFAS), agrochemicals, and other emerging pollutants [8,12,13]. Through adsorption, the bioavailability of pollutants is reduced, minimizing risks to the biotic and abiotic components of the environment, which is the basis for risk-based approaches in environmental remediation. In recent decades, many synthetic and natural adsorbents have been used to remediate a wide range of organic and inorganic pollutants [14–16]. Synthetic adsorbents are suitable for treating contaminated water and soil due to their high surface area and adsorption capacity. Activated carbon/carbonaceous materials, nanomaterials, clay composites, ion exchange resins, composite adsorbents, and polymeric sorbents are among many in that category [17–21]. Natural adsorbents, including agricultural and industrial waste, clays,
biopolymers, and cellulose-based materials, are low-cost and environmentally friendly. These features make them attractive alternatives to synthetic materials if they have the desired adsorption capacity [22–25].

Challenges exist with the development of novel adsorbent materials, including their adsorbing performance, kinetics, stability, selectivity, regeneration, and cost effectiveness. Developing sustainable environmental adsorbents is also an important consideration, i.e., whether the sorbent meets the criteria of being “green” or “eco-friendly”. A “green” or “eco-friendly” adsorbent is a material that can selectively bind or adsorb pollutants or other substances from a solution or gas phase without causing harm to the environment [26], which also aligns with the twelve principles of green chemistry [27]. It is expected to be derived from renewable, biodegradable resources and has a low environmental impact during production, use, and disposal [15,28]. From a material perspective, the technical challenge is to obtain all these properties in a single adsorbent or find a balanced approach that is fit for purpose. For example, if regeneration of adsorbent materials is desirable, the sorption capacity might be compromised to meet this purpose. Therefore, developing novel adsorbents with desirable properties is vital for environmental remediation.

The prospective environmental applications of adsorbent materials are multifaceted and include water treatment and soil stabilization, and interest in their development has grown in recent years [15,29–31]. Low-cost resources, e.g., smelter slag, biochar, red mud, clays, biomass, and their derived materials, such as nanoparticles, have been increasingly used to prepare adsorbents, which is the focal point of this review considering circular economy prospects. This review also focuses on advancements in synthesizing adsorbent materials, explicitly analyzing the assessment of preparation techniques, consequences for ecotoxicity, and potential for reuse or regeneration. Various preparation techniques, including chemical synthesis, physical modification, and bio-based approaches, have been evaluated for their efficiency in improving surface characteristics and adsorption capacity. The benefits and drawbacks of every technique are discussed to offer insights into the most promising strategies for producing adsorbent materials. Additionally, this review looks at the sustainable attributes of potential sorbent materials for environmental remediation, including the toxicity, reusability, and regeneration of the specific sorbent material(s). This understanding can be extended to developing “green” adsorbents for environmental remediation.

2. “Green” or “Eco-Friendly” Adsorbents

A family of materials known as “green” or “eco-friendly” adsorbents has recently attracted much attention because of their favorable effects on the environment and their use in a range of remediation industries [32]. This type of badge on adsorbents has been rewarded based on three primary considerations: (i) the source of the raw material, (ii) the fate and impact of the adsorbent on the applied medium, and (iii) the reusability of the adsorbent. The adsorbent may be derived following one or all of these considerations.

Green adsorbents are frequently made from naturally occurring, easy-to-process minerals or renewable resources, such as bio-based polymers, natural fibers, agricultural waste, and industrial waste [30–32]. Green adsorbents use these materials to lessen their dependency on non-renewable resources and help minimize their carbon footprints. Eco-friendly adsorbents are also characterized by their low toxicity and biodegradability. These adsorbents are made to have as little impact on the environment as possible while efficiently remediating pollutants or toxins from air, soil, water, or other media. Green adsorbents are made to reduce their adverse effects on ecosystems and human health, in contrast to traditional adsorbents, which may degrade and release harmful chemicals into the environment [29,30]. These adsorbents are easily reusable through recyclability with some downgrading of the adsorption properties or after a low-cost regenerative process by which the original properties of the adsorbent can be restored. To improve the performance and adaptability of green adsorbents, researchers are constantly investigating the synthesis
of novel materials. These adsorbents provide a promising way to address environmental issues and encourage a greener and more sustainable future.

Here, we used the term “adsorbents or sorbents” to describe the materials that act in the sorption process, excluding their role as the sole catalyst. A literature search was conducted to analyze papers with relevant words. The results from searching the Scopus database for both “adsorbent” and “remediation” in the article title, abstract, or critical keyword area of the record produced 3623 records from 2017 to the present (search date 1 November 2023). Among them, 2827 original studies were identified with no further filtering or quality sorting. We selected only English-language articles (2781), further filtered “within the results”, and obtained revised records. To do that, we used the following restriction: “(((TITLE-ABS-KEY (“adsorbent”) AND TITLE-ABS-KEY (“remediation”) AND PUBYEAR > 2017 to 1 November 2023)) AND (“eco-friendly” OR “regeneration” “eco-friendly” OR “reusability” OR “ecotoxicology” OR “green” OR “eco-toxicology”) AND (LIMIT-TO (DOCTYPE, “ar”)) AND (LIMIT-TO (LANGUAGE, “English”))”. This specific search identified a total of 2134 articles. We exported these articles into Endnote software (v20.2.1, Clarivate™, London, UK) and carefully checked them for quality. This included removing review articles from the cohort and removing confusing terminology, such as “Malachite Green”, that did not study any “green” or eco-friendly aspects. Finally, 2018 articles were clustered manually into several categories and are presented as a box chart (Figure 1). The number of publications on this topic increased during the past few years (2017–2022), i.e., it increased from 134 in 2017 to 610 in 2022.

![Figure 1](image_url). The relative weight of the key “eco-friendly” aspects studied in original research published between 2017 and 2023. There are a few overlapping subject matters, such as the antimicrobial properties of materials vs. environmentally bio-safe materials. These are also reported in the box plot. The values in parentheses for the reported categories are the number of original papers listed in the Scopus database. Details of the data screening and quality control procedures are provided in the main text.
The box plot shows a significant discrepancy between the adsorbents claiming to be “sustainable” or “eco-friendly” and the standard indices, such as “reusability” (regeneration or recycling) and “ecotoxicity” assessment. Approximately 28% of the 2018 published papers reported reusability data, while only 1.5% reported ecotoxicity assessment. Both indices were left out almost entirely.

It is difficult to verify the degree to which the adsorbent is “green/eco-friendly” by learning only the reported outcome. A comprehensive assessment of its chemical footprint and cost analysis are needed. For example, Feng and colleagues [33] developed a graphene-based nanocomposite that removed drug residues (diclofenac) from water. This adsorbent is deemed to be reusable and non-toxic to the water bacterium Escherichia coli. However, the formulation pathway of the adsorbent requires the following chemicals along with a series of thermodynamic energy inputs: graphene oxide, terephthalaldehyde, tetrakis (4 aminophenyl) methane, 1,4-dioxane, other salts, and solvent. A similar claim was made by Bedadeep et al. [34], indicating the chemical and energy input footprint for the synthesis of their adsorbent seems to be high. The authors emphasized the green synthesis approach, considering the growing demand for reducing toxic chemical usage where possible [35].

This argument does not suggest that one adsorbent is better than another, but the adsorbents should be designed with the least possible environmental impact while still providing the desired performance. In this review, we aim to illustrate the primary considerations when developing “green” or “eco-friendly” adsorbent materials. This includes the use of waste or low-cost materials, environmentally friendly processes for preparation, ecotoxicity considerations, and reusability and regeneration. These considerations are targeted to achieve sustainable development goals and a circular economy (Figure 2).

Figure 2. Various core aspects of the “green” or “eco-friendly” sorbent materials.

3. Use of Waste Materials for the Synthesis of Low-Cost Green Adsorbents

Various low-cost, renewable, and waste materials are being investigated to prepare adsorbent materials, including clays and zeolites, recycled materials, and agricultural and industrial wastes. This is an increasing trend in this research area, as evidenced by the Scopus document search (TITLE-ABS-KEY (sorbent) AND (low cost) OR (waste)). The web search results with keywords “((waste materials) AND (green adsorbents)) AND (green synthesis)” from PubMed data show a significant interrelationship with waste-materials-derived green adsorbents and their utilization for organic and inorganic contaminants (Figure 3). In total, 124 articles were collected to draw the co-occurrence map constructed using the VOSviewer software (V. 1.6.20). The visualization network map indicates the degree of relatedness between keywords. It highlights the research trend of utilizing different waste materials for environmental remediation. Research related to the green synthesis of adsorbents from different waste materials frequently
considered the sorption and degradation of different organic and inorganic contaminants from soil and water (Figure 3). Understanding remediation mechanisms by utilizing spectrometric characterization techniques remains a recent research interest, alongside bringing novel synthesis techniques from different waste materials. The rise in using low-cost waste materials for developing remediation adsorbents is also because governments, environmental regulators, and industries are overwhelmed by the disposal and recycling of these resources [36]. Using waste or low-cost sorbent materials may not reduce waste as much as waste-to-energy or waste-to-building/construction materials approaches can [37,38]. However, it can still provide significant benefits, including reducing waste volume, decreasing environmental pollution, and improving resource efficiency. Waste and low-cost materials can also generate value-added products to provide additional economic and social benefits.

Figure 3. Network visualization maps show co-occurrences of the most used keywords in the “Title/Abstract” search criteria. A total of 124 publications have been selected from 2010 to the present date (14 April 2024) with keywords searched as “(green adsorbent) and (waste materials) and (green synthesis)”. The figure highlighted the research trend with different colors, showing keyword distribution over the highlighted timeframe.

Low-cost, abundant, and degradable precursor materials can be considered for the eco-friendly production of sorbent materials. Such considerations include agricultural and industrial waste, natural and low-cost minerals, and municipal solid waste (Figure 4). The number of raw or waste materials is growing over time due to decades-long research efforts toward sustainable reuse of waste materials including agricultural waste [29–31], shell-based waste [39], red mud and fly ash-based materials [40–43], biosolids [43], and clay minerals [25,43]. Such information is vital for continuously developing sustainable sorbent materials from renewable resources. This review highlighted some examples of potential feedstock materials with their advantages and disadvantages (Table 1 and Figure 4).
Figure 4. Illustration of waste-derived feedstocks for the development of green adsorbents with eco-friendly synthesis methods.

Table 1. Various feedstock materials with their advantages and disadvantages for the synthesis of adsorbents [44–55].

<table>
<thead>
<tr>
<th>Sources of Feedstock Materials</th>
<th>Examples of Feedstock Sources</th>
<th>Advantages</th>
<th>Disadvantages</th>
</tr>
</thead>
<tbody>
<tr>
<td>Agricultural waste</td>
<td>Husks, straw, cottonseed hulls, crop leftovers, sugar beet pulp, grape bagasse, fruit peel, tea waste, green coconut shell, pine bark, sawdust, wood chips, nutshells</td>
<td>Abundance, Low-cost, Waste management, Renewable and eco-friendly, Carbon sequestration</td>
<td>Variability, Preprocessing requirements, Limited adsorption capacity, Pollutants interference, Design challenges</td>
</tr>
<tr>
<td>Animal wastes</td>
<td>Manure, feathers, hair, bones, shells</td>
<td>Resource availability, Cost-effective, Waste management, Carbon storage</td>
<td>Odor and health concerns, Pollutant presence, Variable composition, Low adsorption efficiency and stability</td>
</tr>
<tr>
<td>Low-cost mineral resources</td>
<td>Clay minerals (e.g., palygorskite, smectite, halloysite, kaolinite), other non-argillaceous minerals like oxides (e.g., goethite), slag, diatomaceous earth, zeolite</td>
<td>Abundance, Adsorption capacity, Natural and sustainable, Scalability and stability, pH stability</td>
<td>Limited selectivity, Regeneration, Slow kinetics, Variability, Impacted soil chemistry</td>
</tr>
<tr>
<td>Industrial wastes</td>
<td>Red mud, fly ash, organophosphate, mine tailings, blast furnace sludge/slag/dust</td>
<td>Waste utilization, Sustainability, Tailorability, Large-scale production, Cost-effectiveness</td>
<td>Pollutant transfer, Variability and processing challenges, Regulatory compliance, Limited availability</td>
</tr>
<tr>
<td>Municipal solid wastes (MSW)</td>
<td>Food and green waste, glass, metals, plastic, paper/cardboard, rubber and leather, wood, waste, etc.</td>
<td>Abundant availability, Waste minimization, Tailoring adsorbents, Waste valorization, Zero waste policy implementation</td>
<td>Pollutant content, Inhomogeneity, Additional pretreatment, Odor and aesthetics, Regulatory compliance</td>
</tr>
</tbody>
</table>
3.1. Agricultural Waste/Biomass

Agriculture byproducts have been considered the most significant feedstock sources [56], which provide enormous opportunities to develop various remediation agents. Various agricultural wastes/biomass have been studied to develop novel adsorbents that can remove organic and inorganic pollutants. Compared with conventional adsorbents, agricultural waste is frequently accessible at little or no cost, which decreases the overall cost of contamination cleanup [57–59]. Due to affordable cost and ambiguous availability of agricultural waste, it is suitable for use in underdeveloped or developing nations or places with limited financial means [60–64]. Using agricultural waste as an adsorbent can help lessen the dependency on nonrenewable resources. The circular economy and sustainable development concepts align with this strategy, reduces resource efficiency and environmental impacts (Table 1).

These materials contain various fibers, hemicellulose, cellulose, lignin, ash, biopolymer, and moisture. These functional constituents largely determine their utilization as sorbent materials. For example, natural biopolymers, such as chitosan-based adsorbents, are biocompatible and ecologically friendly, making them valuable alternatives for preparing sorbent materials. Chitosan-ethylenediaminetetraacetic acid-modified biochar [65,66] and rubber-seed-shell-based activated carbon [67] have been reported for the remediation of heavy metal(loid)s in soil and water in addition to CO$_2$ capture. Starch and carbohydrates are two primary components of different food waste. Starch is one of the most abundant and naturally occurring polymers that can be used to prepare sorbent materials. In addition to that, carbohydrates constitute glucose units and glycosidic linkages and possess active and replaceable hydroxyl groups that can be readily cross-linked with other functional groups. Tailoring their functionality with different physical and chemical treatment could also contribute to potential remediation agents [68]. For instance, Li et al. prepared a sorbent—iron-modified bacterial biomass—to remove antimony (III), and they argued that it had a higher sorption capacity than certain previously reported sorbents [69]. In addition, Zhang et al. produced biochar from wood pyrolysis under oxygen-limiting conditions (650 $^\circ$C for 4 h) to immobilize cadmium (Cd) and copper (Cu) from soil [70].

The high surface area, porous structure, and functional group content of agricultural waste make it an effective adsorbent. These properties improve its capacity to adsorb impurities from soil and water matrices, such as organic pollutants, heavy metals, PFAS, agrochemicals, and dyes [57,59,60,71–74]. Adsorbents from agricultural waste often exhibit low toxicity, reducing the likelihood of secondary pollution [75,76]. Furthermore, their use as adsorbents can lessen the requirement for energy- and chemical-intensive procedures, resulting in a reduced carbon footprint compared to conventional techniques [77,78]. To optimize the adsorption efficiency of adsorbents made from agricultural waste, several characteristics need to be optimized, such as particle size, pretreatment techniques, pH, and contact time [72,79,80]. It is imperative to standardize both synthesis processes and characterization methodologies to guarantee uniform remediation performance among diverse waste types and pollutants. Agricultural waste can have different compositions depending on several variables, including crop variety, region, and harvesting technique. This variability may impact the adsorbent capacity and effectiveness, requiring careful characterization and batch-to-batch quality control. Adsorbents made from agricultural waste can effectively remove pollutants but face challenges in their regeneration and reusability due to poor thermal and chemical stability [81]. Thus, the long-term stability of agri-waste-derived adsorbents should be prioritized during the development of adsorbents. More investigations and technological developments are needed to expand the use of agricultural waste as adsorbents from the laboratory to industrial or field-scale applications. Practical implementation requires large-scale manufacturing and deployment. Using agricultural waste as a green adsorbent for the remediation of pollutants has many benefits, such as its availability, cost-effectiveness, renewability, and minimal environmental impact [13,82,83]. However, for this strategy to be implemented successfully, the issues of uniformity, waste variability, regeneration, and scale-up need to be addressed (Table 1). Further study and
cooperation among scientists, engineers, and policymakers are required to fully realize the potential of agricultural waste as a sustainable solution for pollutant remediation.

3.2. Animal Wastes

Feathers, hair, manure, bones, and shells are among the most investigated animal wastes for developing sorbent materials [84–89]. The keratin protein in feathers and hair, organic matter and nutrients in manures, and calcium and phosphate minerals in animal bones can be chemically or thermally modified to prepare sorbent materials [90,91]. Nitrogen, phosphorus, potassium, ammonia, organic matter, carbon, sulfur, and trace minerals (e.g., copper and zinc) are the major chemical components of animal waste [16,90,92,93]. However, the chemical composition varies depending on the type of manure and the food habits of particular animals [94]. These components are essential for nutrient cycling and agronomical benefits in soil systems. However, the raw or aerobic degradation products of manure applied to agricultural soil might cause carbon dioxide emissions due to the low stability of carbon. Thus, understanding the chemical properties of individual animal waste is essential for sustainable waste management. In addition, studying manure waste with advanced characterization techniques provides insight into animal diets, animal health, and overall management practices [95,96]. Transforming animal waste into a valuable resource lowers the need for disposal and associated environmental risks, benefiting waste management. Adsorbents from animal feces have good adsorption qualities because of their large surface area, porous structure, and organic matter content [86,97]. These properties are useful to efficiently immobilize pollutants from water and soil matrices, and they can be reactive to a range of pollutants, including organic pollutants, heavy metal(loid)s, antibiotics, and other emerging pollutants [86,97–99]. The adsorbent and pollutants interact physically and chemically during adsorption, which results in the immobilization or degradation of the contaminants [14]. Furthermore, animal dung can be an adsorbent to lessen adverse and unfavorable environmental effects [15]. Adsorbents derived from animal waste are frequently biodegradable and pose negligible ecosystem hazards [14,100]. More investigations and technological developments are needed to expand the use of animal waste as adsorbents from the laboratory to field-scale applications.

3.3. Mineral Resources

Mineral waste refers to the byproducts and residues generated during the extraction, processing, and utilization of minerals, such as tailings, slags, mine water, and rock waste. If not managed correctly, these materials may contain contaminants or impurities that could impact the environment. On the other hand, clay minerals (e.g., palygorskite, smectite, halloysite, kaolinite), other non-argillaceous minerals such as oxides (e.g., goethite), and diatomaceous earth and zeolite are among the low-cost resources that can also be used to produce sorbent materials. It is worth noting that the cost of sourcing these minerals also depends on the geological deposit and country of the facilities. These minerals’ active surface and manipulable charge behavior provide sorption capacity for various pollutants. Research trends regarding these properties and applications are increasing. The surface areas, porous architectures, and functional groups of minerals contribute to adsorption [101–104]. Removing a broad spectrum of pollutants is one of the main benefits of using inexpensive mineral waste as a green adsorbent [105–107]. Heavy metals, organic pollutants, dyes, antibiotics, and other toxins from soil, wastewater, and groundwater are among them. For example, biopolymer–clay nanocomposites are increasingly investigated for their sorption capacity for a range of contaminants, given the biodegradability and inexpensive and non-toxic properties of biopolymers [108]. Polysaccharides and polypeptides are among the best candidates for the development of biopolymer–clay composites. Low-cost “eco-friendly” resources were also employed when developing clay composites, such as cellulose, starch, chitosan, and peanut hull, given their abundance, biodegradability, biocompatibility, and recyclability. However, including toxicity assay in the investigations is not always the case. Biswas and Naidu [109] utilized an Australian palygorskite clay
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mineral to make functionalized clay sorbent to remove phosphorus from lake water. They argued that this material could effectively bury phosphate ions from lake water. The material did not have a toxic effect on aquatic microorganisms, as demonstrated in the microbial toxicity test.

Depending on the properties of the pollutant and adsorbent, the adsorption may entail physical and chemical interactions such as pore filling, van der Waals interaction, hydrophobic interaction, chemisorption, surface complexation, ion exchange, and electrostatic attraction [110,111]. To improve their adsorption capabilities, they can be readily functionalized or altered using techniques such as heat treatment, chemical modification, or impregnation with organic materials [107,110–115]. Because of their adaptability, adsorbent properties can be tailored to particular pollutants or environmental systems [110,116]. Moreover, recycling mineral waste into useful adsorbents advances waste-to-resource ideas and the circular economy [117–119]. Factors such as regeneration and the possible leaching of adsorbed toxins need to be considered to ensure their long-term viability, and that they present a zero chance of creating secondary pollution.

3.4. Industrial Wastes

Inorganic/organic wastes generated from various industrial processes, including smelting, mining, and mineral refining, have been increasingly investigated for their application as sorbent materials. By converting industrial waste into adsorbents, we can efficiently manage environmental contamination challenges while lessening the disposal burden of tailings [12,120]. Using industrial waste as a green adsorbent for contamination treatment has various benefits [121]. Some examples include fly ash, red mud [84], slag, mine tailings, blast furnace sludge/slag/dust [122], carbonaceous wastes from the fertilizer industry, paper mill sludge, and biosolids from the water industries. For example, the iron and other metallic oxides in smelter slags contribute to the remediation of pollutants [122].

These waste materials contain large specific surface areas, porous structures, and an abundance of functional groups—all of which are intrinsic qualities that facilitate adsorption [121,123,124]. To maximize their adsorption efficiency, the physicochemical characteristics of industrial waste can also be altered by various processes, including activation, chemical modification, and blending with other materials [12,121,125]. Adsorbents obtained from industrial waste have proved to be effective in remediating various pollutants, such as pesticides, heavy metals, dyes, and other emerging pollutants [121,123,124,126–128]. These adsorbents employ a variety of chemical and physical adsorption mechanisms, including covalent binding, surface complexation, ion exchange, and electrostatic attraction [127–129].

3.5. Municipal Solid Waste (MSW)

Municipal solid waste includes food and green waste, glass, metals, plastic, paper/cardboard, rubber and leather, wood, and other wastes [130–132]. These waste materials can be used according to their composition. Current research is almost entirely confined to using thermal energy to convert these wastes into reactive materials such as char or biochar. This pyrolysis can be tailored according to the source materials and desired reactive sites of the sorbent and is an area of increasing interest, given the necessity for a circular economy. The extensive availability of MSWs makes them ideal for use as adsorbents. Since MSW is generated by communities worldwide, it is readily available and might be less expensive than other raw materials [132,133]. The development of decentralized methods for contamination remediation is aided by the local availability of MSW, particularly in areas with potentially inadequate waste management infrastructure [134].

The above-listed materials and feedstocks of adsorbents are not all low-cost, sustainable materials for making sorbents. They are indeed the current lead research topics, with different materials exhibiting different active sites, functional groups, and chemical/physical properties. Continuously exploring the options and providing a case-by-case basis for implementing waste materials for the development of selective remediation agents will achieve sustainable goals and consequently achieve a long-term circular economy.
approach. However, using MSW-based adsorbents presents several issues that need to be resolved. Because MSW is heterogeneous, accurate characterization and processing are necessary to guarantee reliable and efficient adsorption performance [130,134–136]. To ensure long-term environmental advantages and safety, it is also essential to carefully analyze the stability of the materials and the leaching potential over time to understand the possibility of secondary pollution caused by the given materials [136–138].

4. Chemicals and Synthesis Processes

Sustainable and green preparation processes are preferable for the preparation of novel adsorbents because they can minimize the environmental footprint, energy consumption, and waste generation. Examples of the preparation of various kinds of sorbent materials include but are not limited to hydrothermal carbonization methods, microwave-assisted pyrolysis, sol-gel methods, and electrospinning methods (Figure 4). We stress that many raw materials, such as those listed in the “Mineral Resources” section, may be sorbents without additional synthesis or use of chemicals. Here, we will look at methods that are not necessarily green. Yet, our advanced and diverse research expands our knowledge and use of these methods in a green and sustainable way when developing sorbent materials. In this section, different existing synthesis methods are highlighted, along with their advantages and disadvantages for preparing green adsorbent materials, to provide a detailed understanding of each synthesis method (Table 2).

<table>
<thead>
<tr>
<th>Synthesis Methods</th>
<th>Advantages</th>
<th>Disadvantages</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hydrothermal carbonization method</td>
<td>Eco-friendly synthesis, versatility, controlled morphology,</td>
<td>Long synthesis periods, precursor restrictions</td>
<td>[29,139–143]</td>
</tr>
<tr>
<td></td>
<td>energy efficiency</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Microwave-assisted pyrolysis</td>
<td>Increased product yield, reduction of secondary reactions,</td>
<td>Equipment complexity and cost, limitations on heat transfer, feedstock</td>
<td>[144–148]</td>
</tr>
<tr>
<td>method</td>
<td>flexibility, and scalability</td>
<td>uniformity</td>
<td></td>
</tr>
<tr>
<td>Sol-gel method</td>
<td>Tailored surface area and porosity, homogeneity, variety in precursor selection, functionalization potential</td>
<td>Process complexity, uses of reagent, and equipment cost</td>
<td>[149–153]</td>
</tr>
<tr>
<td>Electrospinning method</td>
<td>Tunable morphology and structure, high surface area and pore</td>
<td>Process complexity and scale-up challenges, material compatibility and stability,</td>
<td>[154–161]</td>
</tr>
<tr>
<td></td>
<td>volume, utilization of natural polymers, potential for</td>
<td>cost considerations</td>
<td></td>
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<tr>
<td></td>
<td>functionalization</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Biosynthesis method</td>
<td>Sustainable sourcing, low environmental impact, diverse</td>
<td>Variability in product properties, standardization, quality control, extraction,</td>
<td>[15,29,32,94,143,162–164]</td>
</tr>
<tr>
<td></td>
<td>biomass utilization, biocompatibility and biodegradability</td>
<td>and purification challenges</td>
<td></td>
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</table>

4.1. Hydrothermal Carbonization Method

Hydrothermal carbonization supports biomass conversion into a carbon-rich material by applying simultaneous heat and pressure. It produces “hydrochar” and other carbon materials from a variety of feedstocks, including agricultural waste, sewage sludge, and municipal solid waste [165–169]. Compared to conventional pyrolysis, hydrothermal carbonization is a greener method for synthesizing bio-carbon-inspired adsorbent materials. For instance, conventional pyrolysis methods produce a large amount of toxic syngas (carbon monoxide, hydrogen, and carbon-di-oxide), hydrocarbons (methane, ethylene, propylene), tars, bio-oils, and volatile organic compounds (VOCs) [170–172]. In addition, hydrothermal methods perform at lower temperatures (<300 °C) and pressures than traditional pyrolysis, which may have less of an effect on the environment due to limited energy usage. This phenomenon makes it more environmentally friendly than conventional
pyrolysis [173]. However, conventional pyrolysis methods can also be environmentally friendly because they help to convert biomass waste into valuable products (e.g., biochar or engineered biochar) while reducing greenhouse gas emissions compared to traditional waste disposal methods [174].

Hydrothermal processes can form “hydrochar” with aliphatic structures, amorphous carbon, and mineral contents depending upon the feedstock source and reaction conditions [168]. Inorganic elements, e.g., sulfur (S), calcium (Ca), iron (Fe), magnesium (Mg), phosphorous (P), and potassium (K), can be released from sorbents through hydrothermal processes. This low-cost and eco-friendly hydrothermal method produces high carbon yield and can recover minerals and other inorganic elements depending on the hydrothermal conditions. However, the process may be complex, and carbonization parameters may require optimization. Additionally, potentially hazardous products can be generated, and the scalability of this process might be challenging.

Since water or biocompatible solvents are frequently used in hydrothermal processes, less harsh chemicals are needed, minimizing the chemical footprint and potential risks. These techniques can be used to produce a variety of green adsorbents from a broad spectrum of natural precursors, including organic materials, biomass, and agricultural waste [139,141]. The hydrothermal method is also used to prepare magnetic biochar. The morphology and structure of the resultant adsorbent materials can be altered through hydrothermal synthesis, potentially improving their selectivity and adsorption capabilities [139,143]. By enabling the synthesis process to be conducted at comparatively low temperatures and pressures, this increases the energy efficiency and decreases the overall environmental impact.

Implementing hydrothermal technologies in large-scale production poses some problems despite their effectiveness in the laboratory. Process optimization for adsorbent synthesis, including temperature, pressure, and reaction time, can improve the scalability and efficiency of hydrothermal techniques [141,142]. The variety of green adsorbents that can be produced is increased by selecting appropriate precursors. The scalability and commercial viability of the synthesis process may be impacted by the extended reaction times required for certain hydrothermal reactions.

4.2. Microwave-Assisted Pyrolysis Method

The microwave-assisted pyrolysis method involves rapid heating of biomass using a microwave treatment to produce value-added products. The preparation time can be significantly reduced due to its fast heating and cooling cycles, and thus, the properties of the sorbent can be tailored [147,175–181]. For example, microwave-assisted magnetic biochar has been shown to have up to 10 times greater adsorption capacity, surface area, and pore volume than biochar prepared via conventional methods [63,182]. The process shows high energy efficiency and yield, and reduced emissions, making it suitable for making environmentally friendly sorbents. We understand there are concerns about the equipment cost, safety, and quality assurance for the specificity of the sorbent. However, we also know that using microwaves is an emerging method for sorbent development, and there is a scope for enhancing its sustainability and scalability.

Microwave irradiation of biomass produces quick and uniform heating, which reduces reaction times and increases energy efficiency. Microwave-assisted high-temperature heating can lead to increased product yields [144,178]. However, microwave pyrolysis can reduce secondary reactions and undesirable byproducts, improving control over the pyrolysis process and improving the quality of the biobased products. This process enables manufacturing a wide variety of bio-based materials by providing flexibility and scalability to various biomass feedstocks [183]. Variability in the characteristics and composition of biomass feedstocks can affect the uniformity and efficiency of the materials; hence, careful feedstock preparation and selection are needed. However, there may still be problems with heat transfer inside biomass particles, which could reduce the overall effectiveness of the pyrolysis process [146,184]. Microwaves make it possible to heat biomass quickly
and evenly. High-value adsorbents can be produced from biomass through integrated procedures that combine catalytic conversion methods and microwave-assisted pyrolysis [164,184]. The application of microwave-assisted pyrolysis is a promising approach for effective and sustainable biomass conversion.

4.3. Sol-Gel Method

The sol-gel method has been intensively investigated over the last two decades, resulting in its frequent use for synthesizing inorganic adsorbents such as silica, alumina, or calcium oxide. The sol-gel method involves the hydrolysis and condensation of metal alkoxides in a solvent. It allows precise control of the properties of materials and the ability to incorporate different functional groups onto the synthesized adsorbents [185–187]. However, this method is time-consuming, sensitive to impurities, and in some instances, expensive. The resultant sorbent can be brittle, which limits its durability and reusability. To synthesize adsorbents, a broad range of natural precursors and ecologically acceptable reagents can be used in the sol-gel method, allowing utilization of sustainable and renewable resources [150,152]. The sol-gel process can be used with functionalization to increase the affinity of the adsorbent for target pollutants [151]. By improving the uniformity of the adsorption sites, the sol-gel technique helps to generate homogenous adsorbent [149,152].

The method for synthesizing green adsorbents may require several steps and precise control over the reaction conditions [150]. This can make the process complex and present difficulties for scaling up production. The cost of specific sol-gel precursors and reagents may be greater than that of other materials, and the specialized equipment needed for regulated gel formation and drying may increase production costs [153,188]. The manufacturing of green adsorbents can be made more reproducible and less complex by streamlining sol-gel synthesis through ongoing research into process automation and optimization. Investigating the use of sol-gel techniques to incorporate nanoparticles and nanocomposites could produce advanced adsorbents with improved selectivity and adsorption capabilities. Sol-gel technologies are at the forefront of green adsorbent synthesis for environmental applications despite the cost and process complexity considerations [149,151,153,189].

4.4. Electrospinning Method

The electrospinning method produces nanofibers from a polymer solution using an electric field widely used in science and engineering applications. This method can produce adsorbents with high specific surface area and other desirable properties [156]. Both synthetic and natural polymers are used to fabricate fibrous materials, e.g., polyvinyl alcohol (PVA), polyvinylpyrrolidone (PVP), polyacrylonitrile (PAN), chitosan, and nylon 6 [149–153]. The sorption capacity can be improved by functionalization through modifying or adding fillers to remove pollutants [156]. Further research efforts are needed to optimize the preparation process and ensure the safe application of the adsorbents.

The morphology, porosity, and structure of adsorbent materials produced by electrospinning can be precisely controlled, providing opportunities to modify their adsorption characteristics for particular pollutants [154,155,159]. The interconnected porous structure and high surface-area-to-volume ratio of electro-spun fibers can improve their adsorption abilities and efficacy in eliminating pollutants from a range of media [154,157]. Electrospinning can be applied to natural polymers and biodegradable materials to create environmentally acceptable adsorbents. This approach is consistent with green chemistry and sustainability concepts. Adding particular functionalities to electrospun fibers increases their selectivity for particular pollutants and their overall adsorption performance [156,160].

The electrospinning procedures might require complex setups and precise parameter control, which present commercial application and production scaling issues. The longevity and reusability of the resultant green adsorbents may be impacted by the poor chemical and mechanical stability of specific natural polymers utilized in electrospinning [156,161]. The overall economic viability of electrospinning for large-scale green synthesis can be influenced by equipment cost, energy consumption, and the selection of appropriate
natural polymer precursors [161]. To improve process efficiency and reproducibility, ongoing research has focused on enhancing electrospinning settings, investigating innovative collector designs, and incorporating automation. Adapting the brittle/fragile nature of electrospun adsorbents with high specific surface area offers opportunities to increase their usefulness in various environmental settings, including air purification and wastewater treatment [156,190].

4.5. Biosynthesis Method

Biosynthesis processes involve using natural biochemicals or living organisms to prepare green adsorbents. Here, plant extracts, biopolymers, plant or microbial extracts, and enzymes are considered as feedstock materials for the synthesis of adsorbents. These methods are mainly used for the synthesis of nanoparticles. The green synthesis of iron (Fe)-based, silver (Ag)-based, titanium (Ti), zinc (Zn), and copper (Cu) nanoparticles, as well as bimetallic nanoparticles, has been widely investigated [191–194]. For example, Cynomorium coccineum extract was used for the synthesis of Cu nanoparticles for dye removal [195]. The phytochemicals in plant extracts act as stabilizing and reducing agents during synthesis [196]. Biosynthesis represents a cost-effective and eco-friendly approach for preparing sorbent materials, mainly nanoparticles. However, the use of “green” chemicals such as plant extracts does not readily prove that the synthesized nanoparticles are nontoxic to environmental biota [197]. They need to be tested for site-specific applications [198]. Some of these biosynthesis procedures could be complex, and regulatory challenges may arise when genetically modified organisms are involved in synthesis.

In line with renewable and sustainable resource utilization, green biosynthesis involves the use of natural sources such as plant extracts, agricultural waste, and microbial biomass [143,164]. Compared to conventional synthesis techniques, the synthesis process frequently uses benign reaction conditions, minimizing the use of harsh chemicals and lowering the environmental impact [164]. A variety of biomasses can be used in green/biosynthesis processes to produce a range of green adsorbents with specific characteristics and functions. Obtaining bioactive compounds from natural sources can be difficult and requires a number of stages, which could increase the overall cost of production. Due to variations in natural sources, the properties of biosynthesized adsorbents may fluctuate, making it difficult to guarantee consistent material quality and performance.

5. Ecotoxicity Considerations

The ecotoxicity of an adsorbent depends on its chemical composition, physical properties, and potential leach of toxic substances into the soil and water environment. Although green or eco-friendly adsorbents are designed to be environmentally safe, there are still chances that they pose a risk to the environment if not properly managed or disposed. The sustainable treatment and disposal of sorbent materials after sorption requires further research [199]. Potential leaching and release of sorbed pollutants or sorbent components can occur if the used materials are not adequately treated or disposed. For example, harmful chemicals (e.g., heavy metals, polycyclic aromatic hydrocarbons) might be introduced into biochar materials during synthesis [200,201]. Thus, post-synthesis washing and degradation methods are important for improving the biocompatibility of these materials. Several elements must be considered when evaluating ecotoxicity. These include the physical characteristics, chemical makeup, and potential leaching of harmful substances from adsorbents. It is also necessary to consider the particular organisms being targeted, dose of the adsorbent, and duration of exposure to the environmental systems [202,203].

We understand that the “ecotoxicity assessment” of sorbents is complex, with factors including “the choice of environmental receptors”, “the length of study”, and “the form of sorbents likely to be available or residual” [204]. Most published reports present “quick” or “fit for purpose” data on ecotoxicity by studying the “toxic” effect of the sorbent on only one or two environmental organisms. For example, Biswas and Naidu [70] argued that their synthesized clay composite was safe in water by studying only waterborne
heterotrophic bacterial growth. Although such preliminary ecotoxicity considerations are helpful for developing sorbents, environmental assessments or life cycle assessments of these materials are necessary to comprehend the sorbent fully. This comprehensive assessment includes production processes, cost benefits, sensitivity analysis, $C$ emissions, and toxic risks associated with $C$ usage, reuse, and disposal. The primary goal of ecotoxicity studies is to ensure that the use of green adsorbents for contamination remediation does not have any negative ecological repercussions. Researchers and practitioners can come up with well-informed decisions about the environmental safety and applicability of these materials by performing thorough assessments [202, 205–207].

Indeed, eco-friendly modifying agents are useful for developing functional materials for the sorption of pollutants and preparing sorbent materials. Nevertheless, the use of “eco-friendly” chemicals in the synthesis process may not always be possible in obtaining the desired sorbents. Highly reactive zerovalent iron nanoparticles (nZVIs) could be a good example, where exhaustive chemical-like borohydride is a much more effective sorbent than plant-based extracts [198]. Therefore, it is also prudent to understand the ecotoxic effects of any material intended to be used for water or soils and balance these effects with other considerations, such as regeneration or reusability, so that the disposal of the sorbent has less ecological footprint. To determine whether green adsorbents accumulate in organisms or endure in ecosystems, it is also essential to study the behavior and fate of these substances in the environment [205, 208]. Adhering to appropriate risk assessment is necessary for maintaining low environmental impact of the green adsorbents. The physical structure, chemical composition, and interaction mechanism of adsorbents with pollutants must all be fully understood to facilitate the environmentally friendly objectives [202, 209].

6. Reusability and Regeneration

Regeneration processes enable quick retrieval and reuse of the used adsorbents multiple times without a statistical difference in performance [210]. Reuse and regeneration can be achieved through a technically viable process, e.g., desorption, with little input cost. The regeneration of adsorbents is only considered a vital issue once adsorbents are used as ex situ remediation agents for wastewater. For instance, when nano biochar is implemented for in situ soil management, the reusability of the adsorbents does not work due to the lack of separability of the spent adsorbents. In contrast, reusability studies could be performed on adsorbents for ex situ surface water or groundwater remediation. However, properly managing desorbed pollutants from spent adsorbents is also vital to sustainable environmental management. In this section, the regeneration processes and mechanisms will be critically evaluated based on the progress of the current research.

The main benefits of feasible reusability of adsorbents are cost-effectiveness and lower material disposal requirements. However, achieving such reusability can be challenging, particularly for large-scale applications. The adsorbent may lose some of its original adsorption capacity and overall effectiveness after regeneration due to incomplete desorption, destruction of active sites, or altered properties (e.g., surface area, porosity) [210]. Baskar et al. [76] summarized the main techniques used for the regeneration of adsorbents, including magnetic separation, filtration, thermal desorption and decomposition, chemical desorption, supercritical fluid desorption, advanced oxidation processes, and microbial-assisted adsorbent regeneration. Pros and cons are associated with these regeneration processes. The reusability/regeneration of a sorbent is a good indicator of a sustainable approach to sorbent development. However, highlighting the sustainability of a sorbent’s “reusability/regeneration” may require caution. This is mainly because of the methods used to regenerate sorbent materials, some of which will be discussed.

An investigation of the reusability of green sorbents used for the removal of aniline indicated that the chemicals (e.g., hydrochloric acid and ethanol) used for regeneration are mostly toxic unless fate analysis is performed [211]. Acids, chelators, alkalis, solvents, surfactants, or even water are commonly used for the regeneration of adsorbents via the sorption of heavy metals. In contrast, solvents (e.g., methanol, ethanol, and acetone) were
used for the desorption of organic pollutants (e.g., dyes and pharmaceuticals). Thermal regeneration might induce the loss of adsorbents, the reaction of adsorbate molecules, and the destruction of adsorbent materials, and this process is relatively costly [212]. Magnetic separation provides a simple recovery of materials, but it only works on sorbent materials with magnetic properties [213]. Pollutants can be volatilized or broken down by heating the spent adsorbent to high temperatures, which frees up the adsorption sites for further use. Biological treatments help to react to the spent adsorbent in some specific cases. For example, microorganisms can break down organic pollutants from the surface of an adsorbent, thus freeing the functional sites of the adsorbent. While this could be a spontaneous process of obtaining adsorbent back into action, it only works for biodegradable adsorbates. It also involves slow regeneration processes and might cause fouling in adsorbent pores [210].

Researchers have applied various strategies to overcome these challenges, including modifying the surface properties of adsorbents, optimizing regeneration conditions, developing hybrid sorbent systems, and using renewable and sustainable precursors. The regeneration adsorption-desorption model requires “switching” the properties of the adsorbents [212]. Therefore, novel regeneration processes should be considered based on different sorbent materials and their mechanisms for removing pollutants.

The in situ reusability of applied adsorbents is quite complicated. These qualities, which lessen waste production and encourage effective pollutant removal, support the sustainability and economic feasibility of adsorbent materials. Green adsorbents can be efficiently recovered and reused by using suitable regeneration processes [214–216]. This maximizes their potential for long-term pollutant remediation while minimizing the environmental impact [217,218]. Moreover, post-regenerative chemicals should be managed appropriately to avoid the release of residuals and daughter compounds into the environment.

7. Conclusions and Future Perspectives

This review highlights different techniques applied for synthesizing green adsorbents from a range of feedstock materials, such as natural materials, biomass, minerals, and industrial and agricultural waste, while considering their advantages and disadvantages. The thorough investigation emphasizes the importance of sustainable synthesis methods in minimizing environmental pollution and avoiding the use of toxic or harsh substances. This critical description is an effective source for scientists and industry professionals working on environmentally friendly adsorbents for contaminated site management.

It is essential to mention that some generic adsorbents, like biochar or raw earth minerals, can be commercially available and labeled as “green” or “biocompatible.” However, it is possible that there are insufficient publicly available data to back up these claims; they are especially scarce when adsorbents are used to clean environmental contamination. The development of adsorbent materials that are “green” or “eco-friendly” and economically viable remains a research focus for the remediation of contaminated water and soil. These are only possible when we consider not only how to achieve high adsorption capacity but also the sustainability and circular economy of adsorbent materials. Straightforward and practical criterion guidance is needed for the preparation and application of such adsorbent materials. To achieve that, we may find positive outcomes through (1) a consensus definition of green/eco-friendly adsorbents, (2) setting criteria and thresholds for assessing and monitoring the properties, performance, biodegradation, renewability, and environmentally friendly aspects of adsorbent materials, (3) the development of alternative adsorbent materials for industrial needs that are fit for purpose and green/eco-friendly, and (4) regulatory considerations for the choice and application of sorbent materials that are safe for human health and the environment. These combined efforts would benefit the research, industry, and regulatory communities for the future development of green adsorbents and their applications.
Author Contributions: Conceptualisation, Y.L. and B.B.; Formal analysis, Y.L., B.B., M.H. and R.N.; Writing—original draft, Y.L. and B.B.; Writing—review and editing, Y.L., B.B., M.H. and R.N.; Supervision, R.N. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Data Availability Statement: Not applicable.

Acknowledgments: The authors are grateful for the financial support from crc for Contamination Assessment and Remediation and the Environment (crcCARE). B.B. thanks to Alexander von Humboldt Foundation for the fellowship to him.

Conflicts of Interest: Authors Yanju Liu, Bhabananda Biswas and Ravi Naidu were employed by The University of Newcastle and affiliated with the company crcCARE Pty Ltd. The remaining authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest. The crcCARE Pty Ltd. had no role in the design of the study; in the collection, analyses, or interpretation of data; in the writing of the manuscript, or in the decision to publish the results.

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