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Review Article

Agro-Waste as a Potential Source for the Development of Adsorbents in Water and Wastewater Treatment

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Abstract - *Clean and safe water is critical to preserving the Earth's rich biodiversity. Water pollution has become a serious problem worldwide. Materials from agricultural waste are known to be rich in functional groups such as carboxyl, thiol, amide and hydroxyl groups, which enable the binding of pollutants to the surface. Taking advantage of the compositional properties of agricultural wastes, the researcher continues to explore the development of low-cost adsorbents that utilize various available agricultural wastes in water and wastewater treatment as a fascinating field. The article summarizes current research on developing adsorbents, utilizing readily available agricultural wastes to adsorb a wide range of water pollutants. Additionally, this article attempted to explain the optimization procedures, modification strategies and factors influencing adsorption capacity. The literature review revealed that agricultural waste adsorbents have equivalent adsorption capacity to conventional adsorbents. In most cases, surface modification treatments result in an obvious improvement in absorption capacity.*

Keywords - *Sorbent, Adsorption, Agro-Waste, Eco-friendly, Modification, Pollutant.*

1. Introduction

Fresh water is considered a precious resource on Earth due to its limited availability and crucial role in supporting rich biodiversity. [48] Worldwide, vast arrays of diverse pollutants are being introduced into the environment by means of exponential population growth and various industrial activities. According to WHO 2020, over 2 billion people lack access to safe and affordable drinking water, and more than 57% of the world's population expects to have difficulty accessing safe and drinkable water by 2050. Therefore, monitoring and managing contamination of freshwater ecosystems and all alternative sources, including wastewater, must be continued, developed and improved. [48] Many treatment techniques, including chemical precipitation, ion exchange, membrane separation, chemical oxidation, biological degradation, solvent extraction, and adsorption, are available to remove pollutants from aqueous solutions. However, they are not as useful or effective in all societal segments due to limitations like specificity, the production of large amounts of toxic sludge, and high capital costs. However, on the contrary, the adsorption method is considered one of the most effective among all treatment techniques due to its simplicity, easy operation, affordability, adaptability, and high absorption capacities. [1,2,4]

Various materials, including charcoal, clay, zeolite, polymer materials, silica, graphene, nanotubes, and nanoparticles, are used and developed as adsorbents in water and wastewater treatment. Scientists have recently focused a great deal of attention on using materials of biological origin to remove pollutants because functional groups such as amino groups, carboxyl, hydroxyl, and amide on their surface aid in the biosorption process. [4] The physicochemical properties of these waste materials make them a very good adsorbent for removing pollutants such as dyes from industrial wastewater and can replace the available, costly commercially available activated carbon and other materials. [1] Harvesting, post-harvesting and processing generate large amounts of agricultural waste worldwide each year, and the inefficient conversion of these resources into other useful pathways results in serious environmental pollution. [2] After processing, the leftover parts of plants, such as broken and damaged grains, husks, leaves, stems, roots, and outer skin, are called agro-waste. [3] The use of agricultural wastes or by-products, however, is crucial to treating wastewater because it offers the added benefit of managing agricultural waste while also treating water. Researchers have successfully used agriculture-based biomass for the removal of various types of pollutants from aqueous solution, considering (i) abundant availability, (ii) low cost, (iii) good adsorption capacity, (iv) ease of physical/chemical modification, (v) easy regeneration, (vi) limited disposal problem, (vii) agro-waste management, and (viii) eco-friendly. Agricultural waste was reviewed as a biosorbent for heavy metal ions by Shaukat et al.2022, and for heavy metal and synthetic dyes by Hina Khatoon and J P N Rai 2016. Researchers have investigated a range of agricultural wastes as an inexpensive adsorbent for removing a wide range of pollutants; their findings are compiled in Table 1.

2. Conventional Treatment Methods

Various physicochemical and biological treatments are developed and used to remove various pollutants, including oxidative remediation, adsorption, membrane separation, coagulation, ion exchange and chemical methods (ozonation, Fenton oxidation), electrochemical oxidation, ultrasonic chemical oxidation and radiation oxidation.

2.1. Reductive and Oxidative Processes

This is a viable and quick process for eliminating unwanted chemicals from water without creating secondary pollutants. However, this method has the disadvantages of incurring high capital costs, having an undefined reaction path, and leaving reductive and oxidative residues in the water that must be removed in the case of hydrogen peroxide. [7]

2.2. Membrane Separation

Membranes are barriers or selective barriers that let certain substances pass through while blocking the passage of others. Advanced membranes are designed and developed in a variety of forms, such as reverse osmosis (RO), microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and selective membranes. [7] Additionally, surface charge and membrane pore sizes are important factors in the selective removal of pollutants. High automation membranes are being designed, but energy consumption is high, particularly in processes where pressure acts as the primary motivator. [7] Even though they have some encouraging benefits, their commercial application is limited by the associated financial burden, membrane clogging, instability at higher pressures, and disposal issues. [11]

2.3. Ion Exchange Methods

Interchangeable functional groups of the resin are replaced by the pollutants to be removed until saturation is reached. An ion exchanger is a solid material that can exchange either cations or anions from the environment. Both cationic/anionic dyes, inorganic ions and metal ions can be removed. [7,11] Although a wide range of ion exchange materials have been designed and developed, they are still not widely used due to high cost and ineffectiveness. [11]

2.4. Coagulation

Some active ingredients known as coagulants, which convert the pollutants into a solid or semi-solid form, are added. Coagulants (mainly aluminium/iron salts) effectively and efficiently remove direct dyes. However, this method is less efficient for acid dye removal, and the associated cost, large sludge production, and inability to quantify coagulant dosage limit its large-scale applicability. [11]

2.5. Electrocoagulation

Wastewater and industrial waste effluents are treated using this method. It can eliminate contaminants that are challenging to eliminate through filtration or chemical treatment techniques, such as suspended solids, emulsified oil, petroleum hydrocarbons, refractory organics, and heavy metals. Electric current is passed through the aqueous solution, neutralizing the oppositely charged ions in the water and causing them to precipitate. [7] Its application was restricted by the corresponding high energy consumption, high capital cost, and production of large chemical sludge (secondary pollutants).

2.6. Photocatalysis

A catalyst that can generate electron-hole pairs when light is absorbed is used. Semiconductor materials are therefore used. Photocatalysis seems to be a good prospect for environmental management, but extensive research is still needed to increase its efficiency and validate large-scale applications. The main limitations are a lack of sunlight sensitivity and lower photocatalyst efficiency. Many metal oxide nanoparticles, such as TiO, AgO, etc., are used to degrade dyes effectively.

2.7. Bioremediation

In this process, harmful contaminants are broken down, eliminated, reduced, detoxified, or captured by microorganisms such as yeast, bacteria, algae, fungi, or algae in either anaerobic or aerobic conditions. [13] However, the assimilation of heavy metal ions like mercury in the food chain may worsen the situation. [12] Additionally, the treatment of water also involves a longer duration. [9]

2.8. Adsorption and Bio Adsorption

This is the physical-chemical interaction between the surface of the solid material and the solute present in the solution. The solid is called an adsorbent, and the dissolved substance is called an adsorbate. This technique has a simple structure and operation, which allows the treatment of large volumes of water at an acceptable cost and time frame.

Since it is a surface phenomenon, the surface and pores of the adsorbent play a key role in the adsorption mechanism. [7] Numerous substances have been developed and used, including graphene, polymer materials, charcoal, silica, clay, zeolite, nanoparticles, and nanotubes. Because the adsorbent has regenerative and reusable qualities, it works well compared to other methods. Scientists are paying close attention to and are particularly interested in materials with a biological origin known as bio adsorbents.

3. Agricultural Waste as Low-Cost Adsorbent

Large amounts of agricultural waste are generated during harvesting, post-harvesting and processing and converted into various valuable products. The use of agricultural waste as an adsorbent plays a dual role: waste management and water treatment. [1] They have a higher potential for removing pollutants due to their physicochemical characteristics, which make them an excellent adsorbent. [1] Below is a list of some agricultural wastes studied by many researchers, either in their natural form or activated form as low-cost adsorbents for water and wastewater treatment.

3.1. Pomegranate Peels

Pomegranate is a fruit rich in polyphenols, flavonoids, anthocyanins, phenolic compounds, gallic acid and tannins. [33] Pomegranate peel is disposed of as an agro-industrial waste in both the juice and home industries. [33] Numerous researchers have employed this waste as an inexpensive adsorbent to remove a variety of pollutants, including phenol MB, MO, and Ni (II) [9], as well as Cu (II) [10] and ammonium. [32]

3.2. Apricot Seed Shell

Prunus armeniaca L., or Apricot, belongs to the Rosaceae family and is extensively grown in Europe, Asia, and North Africa. A significant amount of apricot seed shells are produced during the processing of fruits and seeds, either disposed of as agricultural waste or added to improve soil fertility. [14] The primary components of these agricultural wastes are cellulose, hemicelluloses, and lignin. Activated carbon and the powdered form of this agricultural waste are used to remove various pollutants. The powder form of this material was used to remove acid blue 193 dye [14], in activated carbon form to remove metals like Mn2+ and Zn2+ [22], and similarly in biochar form to remove pesticides like atrazine. [23]

3.3. Barley Straw

It is made up of lignocellulose, which can be used as a starting material to produce ethanol. It is regarded as agricultural waste and is utilized to create inexpensive adsorbents and remove Cu (II) ions from aqueous solutions following citric acid treatment. [31] Cetylpyridinium chloride, a surfactant, is added to modified barley straw and used in aqueous solutions to remove reactive black 5 and acid blue 40 dyes. [30] Strong binding to negatively charged ions or dyes is made possible by the surface being positively charged after being impregnated with Cetylpyridinium chloride. [30]

3.4. Banana Peel

Globally, bananas are grown in the tropics and subtropics. Banana peels are thrown away as agricultural waste in homes and the food industry. Numerous studies have been conducted on using powdered dried banana peel as an adsorbent to capture different types of pollutants, such as methylene blue and malachite green. [27] Banana peels with activated carbon were studied for their ability to remove the dye rhodamine [27], while thermally treated banana peels were found to have a 100% uptake capacity in removing heavy metals like Chromium, Copper, Lead, and Zinc. [28] Analogously, 90% absorption capacity was noted for eliminating pesticides, specifically atrazine and ametrine. [29]

3.5. Rice Husk

Rice Husk is a readily available agricultural waste produced during the milling process. 700 million tons of rice are produced worldwide each year, with rice husk making up around 20% of the grain's weight. [19,20] In most parts of the world, the industrial application is still very small and is limited to energy production, animal bedding, and waste disposal. [19] The contents of rice husk include moisture (10–15 percent), celluloses (25–35 per cent), lignin (26–31 per cent), hemicelluloses (18–21 per cent), and silica (15–17 per cent). [20] It was considered a low-cost and efficient adsorbent for removing heavy metals, dyes, phenol, organic compounds, pesticides, and inorganic anions. [21] Rice husk has been utilized as an adsorbent to remove Pb (II) and Cr (VI) ions [19], cationic dyes like crystal violet and methylene blue [24], and activated biochar to remove ranitidine. [25]

3.6. Orange Peel

Oranges are among the many citrus fruits that are widely available in India. Oranges are produced in 60 million tons annually, whereas 32 million tons of orange peel waste are produced annually. [35] One type of biowaste produced in the fruit and household industries is orange peel. Its primary constituents include lignin, cellulose, pectin, hemicelluloses, chlorophyll pigments, and other low molecular weight substances like limonene. [34] It has been studied how well orange peel absorbs acid violet 17 from aqueous solutions. [36] Orange peel also exhibited good absorption qualities for Ni (II), Cu (II), Cd (II), and Zn (II) ions, as well as high selectivity towards Pb (II) ions. [37]

3.7. Sugarcane Bagasse

Sugar cane (Saccharum officinarum) is grown in large quantities worldwide. Around 1.6 billion tonnes of sugarcane are produced annually, generating around 279 million tonnes of sugarcane bagasse worldwide. [40] Brazil is the world's largest producer, with an annual production of 7,39,300 tons, followed by India with 341,200 tons. [38] Agricultural waste consists of cellulose (44%) and hemicellulose (28%), lignin (21%), ash (5%) and extract (2%) [38]. Researchers around the world are paying great attention to the utilization of these agricultural wastes for various applications, such as the production of biofuels and adsorbents for water and wastewater treatment. Chemically modified sugarcane bagasse is successfully used to remove Cu (II), Cd (II) and Pb (II) ions. Sugarcane bagasse was used to remove nitrite ions with an efficiency of 90%, while sugarcane bagasse biochar was more efficient for Pb (II) than orange peel biochar. [39] A summary of various agricultural wastes for the removal of various pollutants is summarized in Table 1.

4. Modification/Fabrication of Agro-Waste Adsorbents

The literature has reported that the adsorption capacity of raw agricultural waste materials for pollutants is relatively lower due to their inherent chemical composition and structure. [42] As a result, numerous initiatives are being made to modify the surface to increase the adsorption capacity. Chemical and physical surface modifications are being applied to adsorptive materials to enhance their adsorption capabilities. Protonation, amine grafting, metal or metal oxide impregnation, and surfactant modification are examples of chemical surface modification techniques.

Table 1. Summary on utilization of low-cost agro-waste adsorbents for removal of various contaminants

S. No	Agro-waste adsorbents	Pollutants removed	Uptake capacity	Reference
1	Pomegranate peels (Dried Powder)	Phenol, MB, MO, Ni (II)	98 %,	8
2	Pomegranate peels (Dried Powder)	Cu (II)	30.12 mg/g	10
3	Pomegranate peel powder	NH4	88 %	32
4	Apricot seed shell powder	Acid blue 193 dye	41.17 mg/g	14
5	Apricot seed shell-activated carbon	Mn (II) & Zn (II)	Mn (II) > Zn (II)	22
6	Barley straw	Nitrate	80 %	15
7	Modified barley straw	Cu (II)	88.1 %	31
8	Banana peel	Nitrate	80 %	16
9	Banana peel	Malachite green (MG) and Methylene Blue	Up to 100 %	26
10	Thermally treated banana peel	Cr (II), Cu (II), Pb (II), and Zn (II)	100 %	28
11	Activated carbon of banana peel	Rhodamine B	81.9–85.6 %	27
12	Used black tea leaves	Basic Violet 3 Dye	345.7mg/g	17
13	Neem leaf powder	Fluoride	80 %	18
14	Rice husk	Pb (II) & Cr (IV) ions	Pb (II) > Cr (IV)	19.
15	Rice Husk	Methylene blue and crystal Violet	24.48 mg/g & 25.46 mg/g,	24
16	Rice husk-activated biochar	Ranitidine	88.3 %	25
17	Orange peel	Ammonia & Nitrate	100 %	34
18	Orange peel	Acid violet 17	87 %	36
19	Orange peel	Pb (II) is selective but also adsorb Ni (II), Cu (II), Cd (II) and Zn (II) ions	32.507mg/g	37
20	Sugarcane bagasse	Cu (II), Cd (II), and Pb (II) ions.	92.6, 149.0, and 333.0 mg/g	39
21	Sugarcane bagasse biochar	Pb (II)	86.96 mg/g	39
22	Chemically modified jackfruit leaf powder	Pb (II)	87.22 mg/g	41
23	Chemically Apple peel	Arsenate, arsenite, chromate, phosphate anions.	15.64, 15.68, 25.28, 20.35 mg/g)	43
24	Potato peel	Reactive Black 5	85.5 %	45

On the other hand, physical modification techniques involve heating materials to remove contaminants and volatile matter. The various modification strategies are presented in Figure 1. Agricultural wastes undergo surface modification processes that change their physicochemical characteristics, including their surface area, porosity, reactive functional groups, bulk and particle densities, and thermal stability, thereby improving the wastes' adsorption performance in terms of removing pollutants. [4, 42] Furthermore, compared to physical modification techniques, chemical modification typically results in better physicochemical properties in terms of surface area, charge density, and functional groups on the surface for favourable and enhanced adsorption. [42] Apricot seed shell activated carbon and nano carbon was utilized to remove Zn (II) and Mn (II) ions [22], and biochar made from apricot shells has a good adsorption capacity for atrazine. [23] The removal of ranitidine was successfully accomplished using rice husk biochar. [25] A barley straw adsorbent for acid blue 40 and reactive black 5 dye in an aqueous solution has been altered by the surfactant Cetylpyridinium chloride. [30] Pb (II) was eliminated from jackfruit leaf powder using a modified form that included 20% isopropyl alcohol and treatments with tartaric acid and sodium hydroxide. [41] Compared to untreated barley straw, barley straw modified with citric acid demonstrated a significantly higher capacity for absorbing Cu (II) ions. [31] Like this, surface modification can be used to remove different pollutants, particularly anions, by applying metal and metal oxides like zinc, iron, and zirconium. For removal of anions like arsenate (15.64 mg/g), arsenide (15.68 mg/g), chromate (25.28 mg/g), and phosphate (20.35 mg/g), surface-modified apple peel containing zirconium cation was applied. [43] Biochar prepared from apricot shells exhibits good adsorption capacity for atrazine. [23]

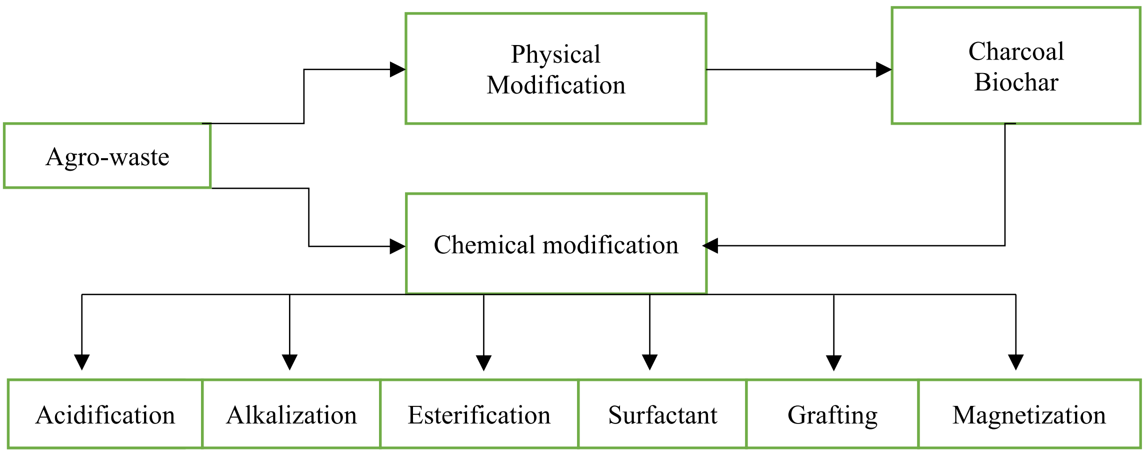

Fig.1 Presentation of surface modification processes

4. Parameters Influencing the Adsorption Process and Adsorption Capacity

The adsorption process involving agricultural wastes as adsorbents depends on the following factors.

5.1. Contact Time

It is the amount of time needed for the adsorbent and adsorbate to come into equilibrium. It is a crucial component of the batch adsorption process and influences both the kinetics and adsorption processes. Adsorption often rises with time until it reaches a maximum known as the ideal contact time. This pattern results from the transference of adsorbate from the solution phase to the adsorbent's active sites until all the surface-active sites are saturated and further no more adsorption occurs. [12, 31] Table 2 provides an overview of how contact time affects adsorption capacity.

5.2. Adsorbent Dosage

The amount of adsorbent determines how many active sites are available, which affects adsorption. [4] While some studies reported a decrease in adsorption with the increase in adsorbent dosage for a particular concentration of adsorbate [25], this could be due to the possibility of agglomeration and congestion of adsorbent particles which will diminish the overall surface area of the adsorbent. [14] In general, most adsorption processes increase with an increase in adsorbent dosage due to an addition in the number of available active sites. **Table 3** summarizes the ideal conditions for the adsorption of various pollutants.

Table 2. Parameters influencing adsorption capacity

Adsorbent	Adsorbate	Adsorbate dosage	Adsorbent dosage	PH	Contact time	Removal	Reference
Apricot seed shell	Acid Blue Dye	20 & 40 mg/l	0.2-2g/L	2-10	0-240 min	41.17 & 24.71 mg/g	14
Rice Husk Activated Biochar	Ranitidine	50 mg	0.5-1.5 g/L	3-11	2-140 min	5.72 - 22.99 mg/L	25
Banana peel	Nitrate	200 mg/l	0.1-25gm	2-6	15-1440 min	86 %	44
Potato peel	Reactive Black 5	50-400 mg/l	0.2-1g	3-11	5-120 min	82.21-95.06 %	45
Sugarcane bagasse ash	Ammoniacal nitrogen	50 mg/l	2-60 g/l	2-12	20-250 min	15-60 %	46
Potato peel Char	Cd (II)	10-200 mg/l	0.1-3g/l	3-9	5-60 min	34.82-98.40 %	47

5.3. Initial Adsorbate Concentration

The driving force behind the uptake of an adsorbate onto an adsorbent's active site is the initial concentration of analyte in the solution. [4] Generally, the adsorption process intensifies as the initial sorbate concentration rises until equilibrium is reached and the adsorption sites are occupied. Moreover, removal capacity decreases as the initial analyte concentration rises, attributed to the saturation of the adsorbent's active sites. [14]

5.4. Effect of pH

An important factor in the adsorption process is pH. It influences the electrostatic interaction between the adsorbent and adsorbate, the surface charge, the solution chemistry, and the adsorbent's surface functional groups, all of which have an impact on the uptake capacity. It is crucial to ascertain the pH at zero charge (pHpzc) to comprehend how pH affects adsorption. The pH point of zero charge (pH_{PZC}) determines the amount of uptake for charged analytes. [14] When the pH of the solution is higher than pHpzc, the adsorbent surface becomes positively charged, which promotes the uptake of anionic ions. Conversely, when the pH of the solution is lower than pHpzc, the adsorbent surface becomes negatively charged, favouring the uptake of cationic ions. [14, 19]

5.5. Surface Area

The adsorption process is a surface phenomenon; therefore, the sorbent's surface area also plays an important role in the adsorption process. The adsorption processes increase with the increase in the surface area of the adsorbent and vice versa. All the parameters influence the adsorption capacities of an adsorbent. Therefore, assessing an adsorbent's removal capacity requires careful consideration of its optimization. The optimum conditions of different adsorbents are summarised in **Table 3**.

Table 3. Optimum conditions in adsorption of different agro-waste adsorbents for organic and inorganic pollutants.

Adsorbent	Adsorbate	Adsorbent dosage	Adsorbate dosage	Contact time	pH	Adsorption capacities/ % Removal	Reference
Apricot seed shell powder	Acid blue 193 Dye	20 mg/L	0.2 g/L	120 min	2	41.17mg/g	14
Rice husk Activated Biochar	Ranitidine	1g	50mg/l	140	9	83.3	25
Apricot seed peel activated carbon	Mn (II)	0.5g	5 mg/L	25 min	3-4	82	22
Pomegranate acid activates carbon	MB	1g	100mg/L	120 min	9	86.79	8
Pomegranate acid activates carbon	Ni (II)	1g	300mg/L	120	4	83.02	8
Banana peel	Nitrate	0.1g	200mg/L	120 min	5.5	80	44
Potato peel	Reactive Black 5 dye	1g	50 mg/l	60 min	3	85.5	45

6. Techniques for Characterization of Adsorbent

Characterizing the adsorbent is very important to determine the chemistry, surface structure/composition, surface area, thermal stability, and particle size, which are essential for developing the adsorbent and understanding the adsorption mechanism and processes. Agricultural biomass is rich in cellulose, hemicellulose, pectin, lignin, lipids, proteins, carbohydrates, phenolic compounds, flavonoids, and pigments, which provide many functional groups such as carbonyl, hydroxyl, carboxyl, amide, amino and thiol groups that Binding sites facilitate the analytes. Numerous techniques, including Fourier Transform Infrared Spectroscopy (FTIR), Thermo-gravimetric analysis (TGA)/DTA analysis, Boehm titration and point of zero charge (PHpzc), Scanning electron microscope (SEM) Transmission electron microscope (TEM), elemental analyzer, BET method, and X-ray diffraction techniques, have been used to characterize the biosorbents. [10, 16–25] All methods are displayed in Figure 2.

7. Future Prospect

This article discusses in detail the potential use of agricultural wastes as an inexpensive and environmentally friendly adsorbent for the sequestration of synthetic dyes, heavy metals and inorganic pollutants in water. Still, several gaps need to be filled in and developed further.

Searching for novel multi-functional agricultural waste sorbents is required.

i) Developing cost-effective and applicable fabrication methods is required.
ii) Most of the investigations have been carried out on the synthetic water and uptake assessment of real wastewater systems that need to be investigated.

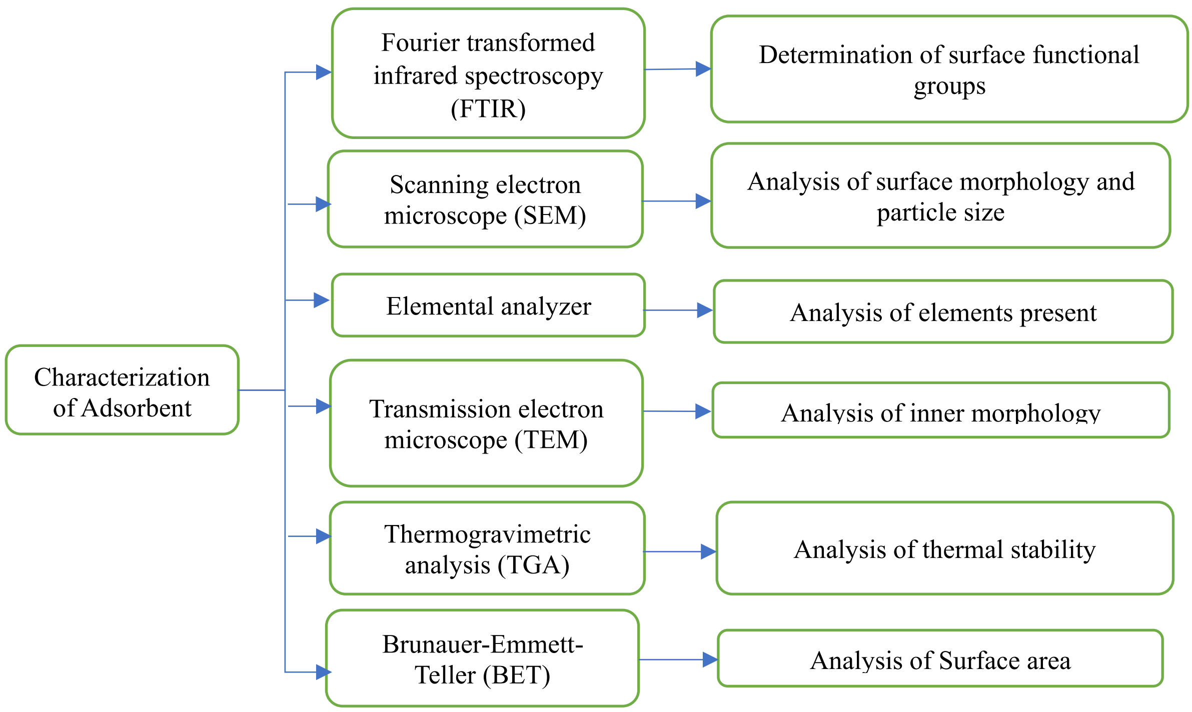

Fig. 2 Characterization techniques and their outcomes.

iii) Development of adsorbents to the commercial scale is a challenge and must be explored for the replacement of traditional adsorbents with agricultural waste-based sorbents as an effective technology in water and wastewater treatment.

8. Conclusion

During the processes of harvesting, post-harvesting and processing, agricultural waste is produced in enormous quantities. This waste is then processed into various valuable products, and using agricultural waste as an adsorbent serves the dual purposes of treating water and managing waste. They could be considered an efficient adsorbent for removing a range of water pollutants because of their physicochemical characteristics and widespread availability. However, raw agricultural wastes have a lower sorption capacity due to their non-reactive physicochemical characteristics. To maximize agricultural waste uptake capacity, surface modifications are therefore required. Researchers can improve adsorption capacities to remove various pollutants by applying various modification techniques to agricultural wastes. The inexpensive, environmentally friendly, locally accessible biosorbent has a greater potential to replace the commercially available inorganic and organic sorbents.

Author Contributions

Mohd Ishaq conceived and designed the review article. He conducted the literature search and compiled and drafted the manuscript. RC Chippa provided critical feedback and participated in drafting the manuscript. Anupama Sharma reviewed selected literature for inclusion in the review. Gh. Ali provided feedback and contributed to the interpretation of the literature.

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