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REVIEW PAPER

Literature review on ability of agricultural crop residues and agro-industrial waste for treatment of wastewater

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Abstract

Purpose Agricultural crop residues (ACR) and agro-industrial waste (AIW) are abundant in Indonesia and primarily used as substitutes for cattle feed or to be naturally decomposed in the nearby environment. This review attempts to examine the potential valorisation of ACR and AIW into biosorbent. This paper also provides the challenges and opportunities in applying wastewater biosorption treatment in Indonesia.

Method A literature review from available literature was carried out to reveal and explore the ability and prospective application of ACR and AIW for treatment of wastewater

Results The reviews show that ACR and AIW can be used for wastewater treatment in different forms including: filter media, activated carbon, biosorbent and biochar. Activated carbon has demonstrated its high ability and efficiency in removing organic pollutants in wastewater. This is due to its large porosity, internal surface area, and mechanical strength. ACR and AIW in wastewater biosorption can be applied in any small-scale agro-industries because of their simplicity procedures, technology, and low cost. Various options of wastewater technologies have also been investigated in recent years. Yet, various issues have been aroused surrounding this technology, including the biosorptive capacity, the performance-effectiveness, the design, and the high operation costs.

Conclusion The study found that problems of a high cost of carbonation and activation process, the needs of regeneration treatment, and the up-scaling or commercialization might hinder the sustainable valorisation of ACR and AIW.

Keywords Agricultural and agro-industrial waste, Activated carbon, Eco-friendly wastewater treatment, Natural biosorbent, Pollutants removal

Introduction

Agricultural crop residues (ACR) and agro-industrial waste (AIW) – characteristics and potential

Agricultural crop residue (ACR) is highly potential in Indonesia, with a continuously increasing trend but limited treatment or conversion technologies are available (Suhartini et al. 2021); either for production of bioenergy or bioproducts (Suhartini et al. 2022a). Therefore, many ACRs are disposed directly to nearby environment or landfill, which can negatively affect the environmental quality and human health (Bolong et al. 2016). The potency of ACR in Indonesia can be seen in Table 1.

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Table 1 The potency of agricultural crop waste in Indonesia

Agricultural crop wastes	Production (million tons)	Refs.
Rice straw	81.90	(Suhartini et al. 2021)
Maize straw	11.50 - 22.90	
Coffee husk	0.38 - 0.45	
Coconut husk	10.40	(Brunerová et al. 2017)
Cacao pod husk	0.61	
Banana peels	2.71	
Rice husk	15.00 - 16.50	(Dhaneswara et al. 2020)
Corn straw	87.5	(Muktiani et al. 2017)
Corn stover (or cob)	5.19	(Ong et al. 2018)
Sugarcane bagasse	9.75	
Rubber wood residues	41.00	(Conrad and Prasetyaning 2014)
Oil palm empty bunches (OPEFBs)	45.86	(Suhartini et al. 2022b)
Soybean straw	0.23	(Krisnawati and Adie 2015; Ministry of Agriculture of the Republic of Indonesia 2018)

Agro-industrial waste (AIW) is any waste material generated from the production process in agricultural-based industries (Mussatto et al. 2012). They found that AIW has now been sought as a potential resource due to its high sugar, mineral, and protein contents, suitable as growing media for microorganisms. Besides, nee'Nigam et al. (2009) reported that AIW contains high cellulose (35 - 50%), hemicellulose (20 - 35%), and lignin (10 - 25%), depending on the type of plant and production process used. In detail, the composition of lignocellulosic waste derived from agricultural and agro-industrial sectors is shown in Table 2.

Table 2 Characteristics of lignocellulosic waste from agriculture and agro-industrial sectors

Waste type	Cellulose (% TS)	Hemicellulose (% TS)	Lignin (% TS)
Barley straw	33.8	21.9	13.8
Corn stover	33.7	31.9	6.1
Corn stalks	35.0	16.8	7.0
Cotton stalks	58.5	14.4	21.5
Oat straw	39.4	27.1	17.5
Rice straw	36.2	19.0	9.9
Rye straw	37.6	30.5	19.0
Soybean stalks	34.5	24.8	19.8
Sugarcane bagasse	40.0	27.0	10.0
Sunflower stalks	2.1	29.7	13.4
Maize straw	32.9	24.0	8.9

Note: TS= total solids. Source: nee'Nigam et al. (2009)

AIW potency in Indonesia is also highly abundant (Suhartini et al. 2021). For example, the total generation of tofu dregs from tofu industries was 1.024 million tons (Suhartini et al. 2022a). The total generation of various AIW from several fruit- and vegetable-based processing industries, including orange peel, seed and segment membrane (1.20 – 1.44 million tons) (Indonesian Statistics 2021a; Marín et al. 2007), pineapple peels (0.128 million tons) (Anwar et al. 2015); cassava peels and fiber (3.81 million tons) (Ratnadewi et al. 2016); and potato peel (0.272 million tons) (Chavez et al. 2020; Indonesian Statistics 2021b). In the case of other industries such as tempeh and tapioca agro-industries in Indonesia, these also generated solid waste or wastewater which contains high organic material potential to be further valorised into high value-added products such as bioenergy, biochemicals, and other bioproducts (De Corato et al. 2018; Ezejiofor et al. 2014; Zhang et al. 2016). Food waste from the fast-food industry also contributes a huge potential for waste disposal. According to Soma (2017), in Indonesia, of the total municipal solid waste (MSW) production of 40.15 thousand tons/year, the proportion of food waste disposed of to landfills has the highest value (63%), which is mainly derived from the food industry. These findings show a huge potential and availability for ACR and AIW in Indonesia.

Wastewater treatment technologies – current and future trends

Agricultural and agroindustry sectors have positively contributed to the Indonesian economy and played a significant role in providing job opportunities for local communities (Suryaningrat 2014). However, many agro-industries, especially small- and medium-scale agro-industries (SMEs), still have problems with their waste, including wastewater and solid waste. Many of them directly disposed of wastewater to the nearby river or water bodies (Tabatabaei et al. 2022). Such practices cause a detrimental impact on the environment and lead to water pollution, water toxicity or event carcinogenic effect to human and aquatic life (Gao et al. 2010; Lourenço et al. 2015); mainly due to high chemicals or high COD concentration in wastewater (Lim et al. 2010; Lotito et al. 2012a; 2012b; Wei et al. 2015). Hence, the adoption of wastewater treatment technology that is cheap, simple to operate, and can effectively remove pollutants in agroindustry's wastewater is critically needed. Various studies have highlighted several wastewater treatment technologies, as shown in Table 3. For example, aerobic granular sludge (AGS) technology is a promising technology for treating textile wastewater (Franca et al. 2015; Gao et al. 2010; Kolekar et al. 2012; Lotito et al. 2012c; Wei et al. 2015). Other than bacteria and protozoa, AGS is composed of extracellular polymeric substances (EPS), which have functional groups (i.e. carboxyl, phosphonate, amine, hydroxyl groups) that provide binding sites beneficial for non-organic pollutant (i.e. chemical dye) biosorption (Gao et al. 2010; Nancharaiah and Reddy 2018; Wei et al. 2015). Fenton oxidation is one of the advanced oxidation processes (AOPs) suggested for the partial or complete decomposition of pollutants in wastewater, resulting in bio transformed products that are less toxic and more biodegradable (Karthikeyan et al. 2011). A combination of photo-Fenton and aerobic sequence batch reactor (SBR) also had similar performance, which significantly remove both organic and inorganic pollutants in wastewater, producing suitable effluents qualities for water reuse (Blanco et al. 2014). On the other hand, several studies have emphasized the efficacy and the benefits of using natural biosorbent in treating wastewater. For instance, Suhartini et al. (2013) demonstrated biofiltration with a combination of natural biosorbent made of M. oleivera seed and natural filter media (i.e. sand, gravel, bamboo sheet, and coconut coir) effectively removed organic pollutants in tapioca wastewater. Benchekor et al. (2018) reported that natural materials such as from shrimp shells (Aristeus antennautus) can be made into a chitin-based material adsorbent, which was potential to treat the purple NR5 dyes. Abdolali et al. (2014) has also shown that converting lignocellulosic waste material into biosorbent can remove toxic metals ion and dye pollutants in wastewater streams. Several studies found that agricultural crop residues (ACR) such as oiltea waste for dyes removal (Liu et al. 2016a), as well as coconut shells (Kumar and Meikap 2014) and coconut husk (Verma et al. 2021) can be used as biosorbent for removing Cr (VI) from wastewater. While Mo et al. (2018) reported that agro-industrial waste (AIW) has potential as a natural biosorbent of organic and inorganic pollutants in water or wastewater streams. These findings indicated that ACR and AIW could be further used in treating wastewater as a natural biosorbent. Grace et al. (2016) has examined that various waste materials, including coconut shell, tea/coffee waste, rice husk, masonry waste, wood waste, and fly ash, can be used in water treatment processes. This paper investigates the potential valorisation of ACR and AIW into biosorbent and scrutinizes the challenges and opportunities of their application in wastewater treatment in Indonesia.

Table 3 Comparison of wastewater treatment technologies from various studies

Technology	Advantages	Disadvantages	Refs.
Activated sludge (AS)	- Simple operations - Highly cost-effective treatment - Limit the formation of secondary pollution - Has a low capital cost - More eco-friendly than chemical treatment (such as chlorination process)	- Inefficient to degrade complex or toxic pollutants - Has several operational problems such as bulking, foaming, sludge settling, and process instability - Has poor performance in nutrient removal	(Ahmed et al. 2017; Guo and Zhang 2012; He et al. 2017; Ju and Zhang 2015)
Aerobic granular sludge (AGS)	- Effective for removing non-organic pollutants (i.e. chemical dyes) - Can be effectively used as a low-cost and alternative biosorbent - A stable dye removal can be achieved (> 90%) - Can promote complete biodegradation of aromatic amine - The efficacy performance was not affected by dye or its breakdown products - It offers compact and cost-effective treatment	- Highly dependent on pH value - pH of 2 is favourable - Several issues are found surrounding process's stability and longer start-up period (10 - 13 months) - Problems with dewatering and digestibility of AGS - Ideal operational condition for higher nitrogen and phosphorous removal has not yet been established - Full-scale application has yet been established	(Franca et al. 2015; Gao et al. 2010; Kolekar et al. 2012; Liu et al. 2016b; Nancharaiah and Reddy 2018; Wei et al. 2015)
Sequence batch reactor (SBR)	- Simple and flexible operation - Only use a single tank for equalization, biological treatment, and secondary clarification - Full-scale operation has been established - Has an improved removal efficacy - Produce effluent with reusable quality - The technology has better process control and design, reducing manpower in operation - Can be applied in small area - Has 60% lower cost than conventional activated sludge process - It offers smaller foot-print area and a low investment cost	- Need higher level of maintenance than the conventional system - Need higher level of sophistication (due to automatization and advanced controls) - Potential plugging of aeration devices during the cycle - Has timing constraints	(Dutta and Sarkar 2015; Fernandes et al. 2013; Santos and Boaventura 2015; Showkat and Najar 2019; Singh and Srivastava 2011)
Sequencing batch biofilter granular reactor (SBBGR)	- Can be operated at high hydraulic and organic loadings with high performance efficacy - Suitable effluent can be generated with only one biological step - Has a low sludge production (i.e. reducing waste disposal cost) - Smaller reactor can be used - Has lower operational cost than conventional biological treatment - No need post-treatment tanks - Simple and flexible operation - Has an increased resistance to wastewater fluctuation	- COD removal (36 - 80%) was halted due to the presence of recalcitrant fraction - A low TKN removal efficiency (< 30%) due to competition of microorganism for oxygen	(De Sanctis et al. 2020; Di Iaconi et al. 2017; Lotito et al. 2012a; 2012b; 2012c)

Continued Table 3 Comparison of wastewater treatment technologies from various studies

Technology	Advantages	Disadvantages	Refs.
Advanced oxidation processes (AOPs)	- More effective and efficient of degrading toxic and recalcitrant organic pollutants - Effective for removing compounds of emerging concern (CECs), such as pharmaceuticals compounds, personal care products, illicit drugs, and pesticides - Some AOPs can be operated under sunlight or artificial light sources - Low investment cost - Can be used as pre-treatment combined with biological treatment - Can be to remove micro-pollutants (i.e. polishing step or quaternary treatment)	- Less applied for disinfection - High operating cost due to chemical and energy demand - Performance's efficiency is highly dependent on type of AOPs, operational condition, and characteristics of targeted pollutants - May produce unpredictable by-products, thus limiting wide application	(Ameta 2018; Bermúdez et al. 2021; Bethi et al. 2016; Deng and Zhao 2015; Luo et al. 2021; Nidheesh et al. 2013; Poyatos et al. 2010)
Fenton oxidation	- Can be used for partial or complete decomposition of pollutants - Effluents are less toxic and more biodegradable - Has a low investment cost - Effective for removing hydrocarbons - Simple and easy operation - Use less toxic reagents - No mass transfer limitation	- Need high concentration of iron (in solution) - the process is highly depended with the presence of ferric ion - Conventional Fenton oxidation method required acidic pH condition, generated iron sludge, and needed high chemicals - Need subsequent effluent treatment before discharge - Need sludge treatment which required high amount of chemicals and labors; hence the process is not economically feasible	(Bello et al. 2019a; Karthikeyan et al. 2011; Tekin et al. 2006; Tony et al. 2012; Yoon et al. 2001)
Combination of photo-Fenton and aerobic SBR	- Significantly removed organic and inorganic pollutants (> 80%) - Effluents are non-toxic and meet the discharge standard - Produced suitable effluents qualities for water reuse - Reduced processing time by 50% compared with single biological treatment - Can completely degrade toxic and recalcitrant pollutants - Has an improved effluent biodegradability - Need less chemicals and energy than conventional Fenton oxidation	- Complex operation - Operation cost is slightly higher	(Blanco et al. 2014; Elmolla and Chaudhuri 2011; Esteves et al. 2016; García-Montaño et al. 2006; Ramírez et al. 2012; Rodrigues et al. 2017)

Studies on the use of ACR and AIW in wastewater treatment

Various studies have reported ACR and AIW in wastewater treatment either as filter media, activated carbon, biosorbent or biochar, detailed explanation below. Table 4 summarized the materials, treatments, metal ions, adsorbent dosage, maximum adsorption capacity, and drawbacks. Other studies on the use of ACR and AIW for removing other pollutants such as nitrate, phosphorous, pesticide, pharmaceuticals compounds, and salinity are shown in Table 5. These findings also confirmed that different treatment or activation methods influence the efficacy performance of ACR or AIW to remove the pollutants in wastewater.

Filter media

Asim et al. (2020) studied ACR (i.e. coconut coir) as filter media in treating Cu (II)-contaminated water. Coconut coir is a natural fiber from coconut husk widely and traditionally used as a hanging basket due to its high-water retention and absorption. The fiber of coconut coir is a potential biomaterial for heavy metal removal. Their study further revealed that alkali-treated coconut coir has five-fold Cu (II) removal compared to untreated coconut coir. This finding indicated that alkali treatment enhances the water-absorbency properties of the adsorption properties of coconut coir, thus resulting in superficial and effective natural filter materials. Liu and Chen (2017) construct a wastewater purification system using sorghum stalks and oyster shells as natural filter media. With the system, the concentration of suspended solids, biochemical oxygen demand (BOD) and NH_4-N were reduced by 95.3, 97.0, and 99.3%, respectively, meeting the tertiary wastewater treatment effluent standards. Furthermore, carbon from oyster shells could absorb phosphorus in wastewater, accounting for 50%. The study also indicated that this low-carbon wastewater treatment system could achieve better wastewater effluent for irrigation purposes. Ghazy et al. (2016) investigated the use of ACR, such as rice straw, date palm fiber, and wood chips of orange trees as biofilter media in treating municipal wastewater. This study aimed to build sustainable and cost-effective wastewater treatment (at a pilot-scale) and reduce ACR problems in Egypt. Their study demonstrated that the reduction of BOD, COD, total N, and total P were in the range of 66 - 89%, 64 - 87%, 45 - 56%, and 32 - 52%, respectively. Biofilter made of date palm fiber was the most effective media in removing organic pollutants in wastewater. The findings confirmed that biofilter from ACR can be a favorable and sustainable option in creating a low-carbon wastewater treatment plant. Rodriguez et al. (2020) examined wastewater treatment using coagulation (i.e. chemical and electrocoagulation) combined with biofiltration. Various filter media were used in their study, including ACR (i.e. pecan shell, walnut shell), wood biochar, and granular activated carbon (GAC). Biofilter media of wood biochar and GAC has a better performance than with ACR media filter. However, the media filter from walnut shell has a significant turbidity removal over 18 h of operation from 16.6 NTU to 0.49 NTU. Whilst a study by Dele-Afolabi et al. (2018) showed that rice husk and sugarcane bagasse could be made into shaped porous ceramics recommended for further application as filtration media, for instance. Again, these studies demonstrated that ACR could be further utilized in wastewater treatment.

Activated carbon

Crini et al. (2019) highlighted that ACR (i.e. bagasse, maize cob, coconut shells, wood, peat) are applicable for converting into activated carbon, and can be used in water or wastewater treatment. Furthermore, their study also indicated that AIWs are potential candidates as a non-conventional biosorbent. Yahya et al. (2015) reported that activated carbon could be made from ACR and AIW, in which the quality can be different based on the porosity, carbon content, and filterability of the raw materials. They further added that the conversions of ACR or AIW into activated carbon are through the pyrolysis process with and without chemical activating agents. There are three routes of activation of carbon active include physical (i.e. carbonization at 400-850 °C, steam), chemical (i.e. wet oxidation), and physico-chemical (i.e. H_3PO_4/steam, KOH/CO_2, $ZnCl_2/CO_2$) (Heidarinejad et al. 2020; Nayak et al. 2017; Pallarés et al. 2018; Yahya et al. 2015). Özsin et al. (2019) reported that ACR (i.e. chickpea husk) could be converted into activated carbon, which is chemically activated using KOH and K_2CO_3. Their study found that the resultant activated carbon can remove Pb(II), Cr(VI), and Cu(II) with the maximum adsorption capacities of 135.8, 59.6, and 56.2 mg/g, respectively. While a study by Ghorbani et al. (2020) showed that activated carbon from sugar beet bagasse (SBB) was a feasible alternative in which it has 50.45% Cr(VI) removal from aqueous solutions at a dosage of 1.49 g/L. The cost-benefit analysis showed a production cost of USD 1.5/kg biochar with 34% of biochar yield. A previous study by Khan et al. (2016) also reported that rice husk-activated carbon, which was chemically activated, was very effective in removing Cr(VI) from aqueous solutions and wastewater systems. Yang et al. (2019) further confirmed that activated carbon made of walnut shells was also suitable for the effective removal of quinoline from industrial wastewater streams. Yahya et al. (2015) reviewed that there are various ACRs have been studied as activated carbon for wastewater treatment, including palm shell, mango peel, palm kernel shell, coconut shell, ground nutshell, cocoa pod husk, corn cob, rice straw, rice hull, sugarcane bagasse, oil palm shell, bamboo, etc. Another study from Kumar et al. (2017) also reported that activated carbon from rice straw effectively removed Cr(VI) from an aqueous solution. Kilic (2020) proved that activated carbon from corn cob could remove color in textile wastewater by 99%, much higher than using the coagulation method with $FeSO_4 \cdot 7H_2O$ or with $FeCl_3 \cdot 6H_2O$ (accounted for 90% color removal).

Biosorbent

Chandane and Singh (2016) reported that adsorbent from soybean hulls was very effective in removing the color of safranin dye in wastewater streams due to its large adsorption capacity. Their study confirmed that adsorbent from soybean hulls is natural, eco-friendly, and low-cost, and can potentially be implemented at large-scale wastewater treatment plants. Another study by Daud et al. (2018) also showed that empty oil palm fruit bunches (OPEFB) can be converted into a low cost biosorbent which could effectively remove the color from natural rubber wastewater. Similarly, Draman et al. (2015) studied that tea waste and peanut shells are feasible to be used as biosorbent to remove lead poisoning (or Pb(II)) in contaminated water. Their study reported that the use of tea waste and peanut shell biosorbent (at 0.5 - 1.5 g) could remove Pb(II) in the range of ~ 89 - 90% and ~74 - 75%, respectively. The study also demonstrated that a longer contact time increased the removal efficiency. A previous study by Srivastava and Sharma (2013) also found that biosorbent from rice husk was effective in removing Cr(VI) and can be used as an alternative of highly cost biosorbent or potential for treating Cr(VI)-rich wastewater streams. Ponce et al. (2021) evaluated the lignocellulosic biosorbents potential from corn husk, rice husk, and sugarcane bagasse for treating the methylene blue dye. These wastes were found to be viable with the removal rate percentage of adsorption as follows 98.5 ± 1.2%, 95.4 ± 0.8%, and 95.7 ± 1.9% for corn husk, rice husk, and sugarcane bagasse, respectively. Furthermore, the rice husk also has capability to remove other pollutants, i.e., chromium and nickel in range of 5 to 100 ppm (or up to 95% sorption from the aquatic medium) (Basu et al. 2019). Other components from rice husk are the rice husk ash. This content could be utilized as the biosorbent for some specific dyes. A study from Dutta et al. (2014) also revealed that rice husk ash (RHA) was prospective to be used as a low-cost natural biosorbent for removing the cationic dye Brilliant Green (BG) from contaminated water. This study found that the RHA biosorbent could remove dye pollutants with a high adsorption capacity (250 mg/g) within a quick contact time of 15 minutes. Similarly, a study from Mor et al. (2016) also proved RHA biosorbent has an effective removal ability to remove phosphate contamination in the wastewater stream. Tejada-Tovar et al. (2022) demonstrated that banana (Musa paradisiaca) peels waste can also be further valorized as biosorbent to eliminate toxic metal ions (i.e. Ni(II)) from aqueous solution. The study reported that with a dosage of 0.678 g and temperature of 55 ºC, more than 87% of Ni(II) could be removed. According to Kumar et al. (2018), Pine (Pinus densiflora Sieb) bark is also considered a potential biosorbent to remove pollutants of 2,4,6-trichlorophenol from water. Various studies have further highlighted the potential of ACR (i.e. oiltea waste) for removal of methylene blue (Liu et al. 2016a); rice straw, kiln dust, and chrome shavings for removal of organic pollutants in wastewater (Nashy and El-Khateeb 2015); rice husk, palm leaf, and water hyacinth for Cu(II), Co(II), and Fe(III) removal (Sadeek et al. 2015); sugarcane bagasse, peels of various fruits, and wheat straw for arsenic removal (Shakoor et al. 2016); citrus peels, sawdust, and carrot residues for zinc removal (Zwain et al. 2014); and grape marc for Red Bemacid ETL removal (Chergui et al. 2019).

Biochar

Shi et al. (2018) confirmed that the novel magnetic biochar from phoenix tree leaves was applicable for treating Cr(VI)-containing wastewater, with a removal efficiency of 85%. Jang and Kan (2019) reported that engineered biochar derived from alfalfa hays effectively removed tetracycline (TC) pollutants in contaminated water. In their study, the resultant biochar was chemically activated using NaOH, causing an increase in surface area of 796.50 m^2/g and pore volume of 0.087 cm^3/g. Such improvement resulted in a much greater adsorption capacity of TC than non-activated biochar. The amount of TC adsorption was 302.37 mg/g, almost similar to that of commercial activated carbon of Calgon F400). Arrebola et al. (2020) also found that activated biochar from agricultural residues could remove methylene blue-contaminated wastewater. A study by Yap et al. (2017) showed that biochar from coconut shells, which are then microwave activated, has a high surface area of 834 m^2/g. This activation process contributes to a high efficiency in removing Cd and Pb in wastewater. Li et al. (2018) reported that biochar made wheat straw and rice husk provide higher precipitation adsorption of Pb^{2+}, with the value of 70.60% and 83.60% of the total adsorption capacity, respectively. While Liu et al. (2019) studied the efficacy of biochar from corn stalk in removing Pb^{2+} in contaminated waste at a pilot scale. The results indicated that his research offers a way to prepare a low-cost and effective industrial adsorbent biochar for heavy metals adsorption. A recent study by Tong et al. (2020) reported that agricultural wastes (i.e. cow manure and wheat straw) can be used for making montmorillonite (Mt)-biochars. These two biochars were then tested for their efficiency in the removal of 17β-estradiol (E2) from an aqueous solution. The results indicated that wheat straw biochar has superior performance than cow manure biochar, with the maximum adsorption capacity of 62.89 mg/g and 41.02 mg/g, respectively.The findings above demonstrated that ACR and AIW have higher potential for further valorization as low-cost biochar for wastewater treatment.

Type of pollutants removed and its mechanism

The biosorption process has great potential to treat many pollutants, including heavy metals, dye, pesticide, and organic pollutants (Michalak et al. 2013). Different mechanisms facilitate pollutants removal because of the complexity of the biosorbent structure (Fomina and Gadd 2014; Michalak et al. 2013). Understanding mechanisms of pollutant removal can be beneficial for the application. The knowledge of mechanisms is significant in choosing the appropriate type of biosorbent and determining the preparation methods and factors affecting removal efficiency (Singh et al. 2020; Yaashikaa et al. 2021). The capacity of biosorbent is determined by the type of the material, surface morphology, surface structure, and the functional group of biosorbent (Noli et al. 2019).

Table 4 The removal performance of metal ions by agro-industrial waste/agricultural residues

Materials	Treatments	Heavy metal ions	Adsorbent dosage	Max. adsorption capacity/percentage removal	pH	Drawbacks	Refs
Olive stone	Chemically modified	Pb(II)	10 g/L	38.02 mg/g	5	- Lignin content increases with H_2SO_4 treatment - The difference in the removal rate between acid and alkaline treatments is up to 15%	(Martín-Lara et al. 2013)
Sugarcane waste ash	Hydrolysis and condensation reaction	Acid orange 8 dye (AO8)	150-200 mg/L	230 mg/g (90%)	5	- Impurities are present in the ash, thus further washing treatment is required - Selection of adequate pre-treatment is essential to reduce operation cost - Needs more in-depth studies for testing its quality as biosorbent	(Rovani et al. 2018)
Brewer’s spent grain	Oxidation process	U(VI)	900 mg/L	297.3 mg/g	4.7	- Non-porous material - Depends on the irregular shape of the surface capacity	(Su et al. 2021)
Uncommon crops (Coffea arabica fruit endocarp, coconut fruit endocarp, Eichhornia crassipes weed (EC), and Guadua angustifolia plant)	Physically treated	Ni(II) and Cd(II)	0.15 g/25 mL	74.31% (Ni(II)) 95.77% (Cd(II))	6.6±0.2	- High lignin content hinders the extraction of biosorbent. - Salts solubilization and another complexity organic extractive may present	(Correa et al. 2012)
Buckwheat hulls	Acid treatment (hydroxylethylidenediphosphonic acid)	Au(III)	10 mg/100 mL	450.45 mg/g	2.9	Needs intensive maintenance on pH and temperature to control its ions exchange	(Yin et al. 2012)
Olive stone	Chemically treated	Pb(II)	10 g/L	36.55%	-	Preferable use alkaline chemical treatment than acid to degrade the lignocellulose materials	(Ronda et al. 2015)
Buckwheat hulls	Physically treated	Au(III)	1.0 g/L	422.52 mg/g	3.5	Adjustment for the initial pH of pollutant is needed to meet the sorption capacity	(Deng et al. 2014)

Materials	Treatments	Heavy metal ions	Adsorbent dosage	Max. adsorption capacity/percentage removal	pH	Drawbacks	Refs
Corn stalk	Physically & chemically treated	Methylene blue (MB) and crystal violet (CV)	0.25 g/L	566.27 mg/g	5-10	Contains high cellulose crystals aggregation may inhibit the efficacy	(Peng et al. 2021)
Corncob	A low temperature hydrothermal method (453 K)	Fe(III)	1 g/L	163.93 mg/g	-	Interaction between Nano-Fe_3O_4 may reduce the pore diameter and volume	(Ma et al. 2015)
Coffee husk	Physically and chemically treated	Pb^{2+} and Cd^{2+}	10 - 500 mg/L	89.6% (Pb^{2+}) 81.5% (Cd^{2+})	2.0-7.0	- Could not operate well under low pH (2.0 – 5.0) - Could not work effectively when the initial concentration exceeded 500 ppm	(Quyen et al. 2021)
Wheat straw	Physically and chemically treated	Cd^{2+}	1 g/L	46.18 g/mg	2-11	Not suitable for pH < 8 and low temperature (278 K or 4.85 °C)	(Zheng et al. 2021)
Peanut husk powder	Physically and chemically treated	Pb^{2+}, Mn^{2+}, Cd^{2+}, Ni^{2+}, and Co^{2+}	6.5 g/L	99% (Pb^{+2}) 62% (Cd^{+2}) 30% (Co^{+2}) 45% (Mn^{+2}) 38% (Ni^{+2})	6	Working at high dose (1- 5 ppm) and low pH (3- 6)	(Abdelfattah et al. 2016)
Almond shell	Physically treated	Pb(II), Cu(II)	1 g/100 mL	9.0 mg/g (Cu(II)) 13.7 mg/g (Pb(II))	5	Preferable use alkaline chemical treatment than acid to degrade the lignocellulose materials	(Ronda et al. 2015)
Corn silk	Physically treated	Pb^{2+}	1 g/L	90 mg/g	2.0 - 6.0	- Low absorption capacity at low pH value - Needs higher initial concentration of heavy metal	(Petrović et al. 2016)
Tomato waste and apple juice residue	Alkali treatment	Pb(II)	0.0125 - 0.1250 g	152 mg/g (tomato waste) 108 mg/g (apple juice residue)	2.0 - 10.0	- Not preferable at pH < 4 - Need more activation through NaOH treatment	(Heraldy et al. 2018)

Materials	**Treatments**	**Heavy metal ions**	**Adsorbent dosage**	**Max. adsorption capacity/percentage removal**	**pH**	**Drawbacks**	**Refs**
Mangifera indica (mango) seed shell powder	Chemically treated	Pb(II)	ATMS dose: 1 g/L, CFMS dose: 0.5 g/L	59.35 mg/g (alkali treated mango bio sorbent/ATMS) 306.33 mg/g (carboxyl functionalized mango biosorbent/ CFMS)	5.2	Not applicable for the lower lead pollutant concentration	(Moyo et al. 2017)
Tea waste	Chemically treated	Fluoride	0.4 - 8.0 g/L	3.83 mg/g (untreated tea) 10.47 mg/g (Tea-Fe) 13.79 mg/g (Tea-Al) 18.52 mg/g (Tea-Al-Fe)	2.0 - 11.0	Could not effectively work at concentration pollutant < 50 ppm and pH > 9	(Cai et al. 2015)
Corn stover	Chemically and physically treated	Fluoride	1 - 100 mg/L	6.42 mg/g	2.0	Could not work well at base pH and low temperature condition	(Mohan et al. 2014)
Activated bagasse carbon (ABC), sawdust raw (SDR), and wheat straw raw (WSR)	Chemically and physically treated	Fluoride	4 g/L	56.4% (ABC) 49.8% (SDR) 40.2% (WSR)	6.0	- pH 6 -7 is the minimum requirement to fulfill the absorption capacity - pH > 7 reduced the efficiency	(Yadav et al. 2013)
Black tea waste	Alkali treatment	Cu(II)	-	43.18 mg/g	> 7.0	Not optimum at acidic pH condition	(Weng et al. 2014)
Spent seedcake of Calophyllum inophyllum (SSCI)	Chemically and physically treated	Pb(II), Cd(II), and Zn(II)	10 g/L	52.63 mg/g (Pb(II)) 51.28 mg/g (Cd(II)) 17.99 mg/g (Zn(II))	9.0	- Could not operate in acidic pH - The absorbent capacity decreased at dosage above 10 ppm	(Adenuga et al. 2019)
Olive pomace and chitosan	Chemically and physically treated	Pb(II)	400 mg	19.86 mg/g	2.0 - 5.5	At pH > 5.5 reduced the removal rate at pH value	(Sayin et al. 2021)
Moringa oleifera seed husk (MOSH) and Moringa oleifera seed pulp (MOSP)	Chemically and physically treated	Acid Blue 9 (AB9) synthetic dye	0.5 g/L	329.5 mg/g (MOSH) 694.2 mg/g (MOSP)	2.2	pH<3.81 (MOSH), pH<6.08 (MOSP) is not suitable for this biosorbent	(dos Santos Escobar et al. 2021)

Materials	**Treatments**	**Heavy metal ions**	**Adsorbent dosage**	**Max. adsorption capacity/percentage removal**	**pH**	**- Drawbacks**	**Refs**
Dead leaves of Prunus Dulcis	Alkali treatment	Acid green 25 dye	2.14 g/L	28.57 mg/g	2	- Needs acidic pH condition - The feedstock supply depends on seasonal	(Jain and Gogate 2018)
Zea mays waste	Chemically and physically treated	Oxybenzene	0.5 - 2.0 mg	>90%	5.0 - 6.0	Needs acidic pH to have an effective absorption rate	(Lakshmi et al. 2021)
Orange peels	Physically treated	Ammonia and nitrate	4 gm	100%	5.5	Effective load is at fresh condition, thus need large storage (i.e. high operational cost) and the feedstock availability	(Dey et al. 2021)
Pomegranate peel and orange juice by-product	Physically treated	Phenolic compounds	0.01 - 0.02 g/mL	93.13% (pomegranate peel) 89.59% (orange juice by-product)	4.0 - 7.0	- Efficiency of adsorption not well works at a pH > 4.75 - The particle size > 0.373 mm reduced the removal efficiency	(Ververi and Goula 2019)
Lemon peel	Chemically and physically treated	Ni(II)	5 g/L	36.74 mg/g	5.0	- pH 5 is the minimum requirement and pH > 5 did not give any differences capacity of absorption - Needs more acid chemicals/solutions as at pH ≤ 2 showed the highest removal efficiency	(Villen-Guzman et al. 2019)
Litchi peel (LP), orange peel (OP), pomegranate peel (PP), and banana peel (BP)	Physically treated	Cd(II)	10 g/L	230.5 mg/g (LP) 170.3 mg/g (OP) 132.5 mg/g (PP) 98.4 mg/g (BP)	5.0	- Not conducive when reached the pH value of 5 - Need acidic pH condition	(Chen et al. 2018)
Pine (Pinus halepensis) sawdust	Physically treated	Cu, Pb	10 g/L	60% (Cu removal) 80% (Pb removal)	5 - 8 (Pb) 7 (Cu)	At pH > 7 reduced the removal rate capacity	(Semerjian 2018)
Cucumber peel	Physically treated	Lead	1.0 g	133.60 mg lead	5.0	At pH <5 reduced the removal rate capacity	(Basu et al. 2017)

Materials	Treatments	Heavy metal ions	Adsorbent dosage	Max. adsorption capacity/percentage removal	pH	- Drawbacks	Refs
Papaya peel	Chemically and physically treated	Pb(II)	100 mg	93%	5.0	- High dose of absorbent is required for effective removal. - It is not economically viable, thus more modification is required to handle this issue	(Abbaszadeh et al. 2016)
Corn cob and chestnut shell	Chemically and physically treated	Pb(II), Cu(II), Cd(II)	0.1 - 0.5 g/L	166.39 mg/g (corn cob modified) 124.84 mg/g (chestnut shell)	5.0	- Biosorbent cannot work well at pH below 5 - The acidic condition did not enhance the binding reaction	(Chen et al. 2021)
Sugarcane bagasse (SB), rice husk (RH), and castor leaves (CL)	Chemically and physically treated	Pb(II), Ni(II)	50 - 300 mmol/L	Pb(II): 1180, 948, 802 mmol/kg for SB, RC, and CL Ni(II): 810, 698, 432 mmol/Kg for SB, RC, and CL	5.0	- RH and SB has lower pore distribution than CL - SB has the lowest absorption capacity	(Saxena et al. 2017)
Wheat bran and modified wheat bran, eggshell powder, calcined egg shell	Chemically and physically treated	Chromium	-	64% (eggshell) 70.19% (calcined egg shell) ;75.89% (wheat bran); 96.96% (modified wheat bran)	-	Needs longer contact time (i.e. 5 hrs) for higher chromium removal	(Renu et al. 2017)
Terminalia catappa shell	Chemically and physically treated	Methylene blue (MB)	0.8 g/L	88.62 mg/g	5.0	- Did not operate well at pH value of 4-5 - The particle size must not exceed of 36 mm	(Hevira et al. 2021)
Fallen leaves of Ficus racemose	Chemically and physically treated	Acid violet 17 dye	3 g/L	45.25 mg/g (untreated biosorbent) 61.35 mg/g (H_2SO_4 activated biosorbent) 119.05 mg/g (NaOH activated biosorbent)	2.0	- Needs acidic pH condition - The feedstock supply depends on seasonal	(Jain and Gogate 2017)
Palm-oil shells waste	Chemically and physically treated	Phenol	> 0.8 g	98%	-	- Particle size > 0.85 mm reduces the phenol removal efficiency - Needs base pH condition (Buhani et al. 2018)	(Sahu et al. 2021)
Leaves and saw dust of neem tree	Physically treated	Chromium	12 - 20 g/L	~99%	1.0	- Needs very acidic pH condition - The feedstock supply depends on seasonal	(Aggarwal and Arora 2020)

Table 5 The removal performance of nitrate, phosphorous, pesticide, pharmaceuticals, and salinity by agro-industrial waste/agricultural residues

Materials	Treatments	Type of pollutants removed	Adsorbent dosage	Max. adsorption capacity/percentage removal	pH	Drawbacks	Refs.
Nitrate removal							
Potato peel (PP)	Physically and thermally treated (i.e. pyrolysis). With and without combination with egg shell (ES) (at a ratio of 1:2, ES:PP)	Nitrate	1 g/L	53.80 - 62.10% (Combined ES:PP)	7.25	- The performance efficacy is depended on the wastewater characteristics - Longer contact time is needed (120 minutes contact time) - May result in other compounds such as mixture of $CaCO_3$ and apatite - High energy consumption for pyrolysis	(Quisperima et al. 2022)
Rice husk residue	Physically and chemically treated.	- Total nitrogen (TN) - Ammonium nitrogen (NH_4^+–N)	20 g/L	- 84% (TN) - 100% (NH_4^+–N)	8.07	- Longer contact time is needed (120 minutes contact time) - High dose of adsorbent was used - Large chemicals consumption for chemical activation (i.e. may increase operational cost) - Potential environmental impact from chemical usage	(Luo et al. 2019)
Fresh orange peels (Fop) **Spent orange peels (Sop)**	Physically treated. Particle size 2.458 µm (Fop) and 3.415 µm (Sop)	- Nitrate - Ammonia	1, 2, 3, 4 and 5 g in each 200 mL	- Nitrate removal: 100% (at addition of 2-5 g) - Ammonia removal: 100% (at addition of 4 and 5 g)	5.5	- Longer contact time is needed (90 minutes contact time) - High dose of adsorbent was used	(Dey et al. 2021)
Potato peels (PP) **Sugarcane bagasse (SB)**	Physically and thermally treated (i.e. pyrolysis).	Nitrate	6 g/L (with contact time 0 - 200 minutes)	7.1 mg/g (or 60.38%) 3.9 mg/g (or 38.52%)	2, 3, 4, 6, 8, 10, and 12	- PP biosorbent was more effective at basic pH (8 - 10) - SB biosorbent was more effective at acid pH (2 - 4) - The process required longer contact time for higher adsorption efficiency	(El-Nahas et al. 2019)
Rice husk	Physically (pyrolysis) and chemically treated.	Nitrate	1 g/L	8.11 mg/g	-	- The making process of biosorbent is complex and expensive - High energy consumption for pyrolysis - Large chemicals consumption for chemical activation (i.e. may increase operational cost) - Potential environmental impact from chemical usage	(Satayeva et al. 2018)

Materials	Treatments	Type of pollutants removed	Adsorbent dosage	Max. adsorption capacity/percentage removal	pH	Drawbacks	Refs.
Sugarcane bagasse	Physically and chemically treated.	Nitrate	5 - 40 mg/L (at 5 mg increment)	100% (at 5 mg/L) 96.90% (at 10 mg/L)	4.0	- Increasing dosage of biosorbent (>10 mg/L) reduce the adsorption efficiency - The process is highly pH-dependent - Large chemicals consumption for chemical activation (i.e. may increase operational cost) - Potential environmental impact from chemical usage	(Schwantes et al. 2015)
Solid olive waste	Physically and chemically treated.	Nitrite ions	5 g/L	- 67.50% (Chemical activation with zinc chloride) - 13.50% (Chemical activation with phosphoric acid) - 11.75% (without carbonization and activation) - 6.75% (carbonized without activation)	4.5 - 5	- The process is highly pH-dependent, which more effective at acid pH - Longer contact time needed (at 60 minutes) - Large chemicals consumption for chemical activation (i.e. may increase operational cost) - Potential environmental impact from chemical usage	(Zyoud et al. 2015)
Phosphorous removal							
Potato peel (PP)	Physically and thermally treated (i.e. pyrolysis). With and without combination with egg shell (ES) (at a ratio of 1:2, ES:PP)	Phosphorous (P)	1 g/L	85.90 - 91.60% (Combined ES:PP)	7.25	- The performance efficacy is depended on the wastewater characteristics - Longer contact time is needed (120 minutes contact time) - May result in other compounds such as mixture of $CaCO_3$ and apatite - High energy consumption for pyrolysis	(Quisperima et al. 2022)
Pomegranate peel	Physically and chemically treated. Commixing with $LaCl_3 \cdot 7H_2O$ (10 mmol) and $NiCl_2 \cdot 6\ H_2O$ (5 mmol) using solvothermal process	Phosphate (PO_4)	0.5 g/L	226.55 mg/g (or 97.14% for 700 minutes contact time)	2.35 - 10.84	- Higher weight lost at temperature >160 ºC - The process efficacy reduced at pH > 7.38 or pH < 4.44 - Large chemicals consumption for chemical activation (i.e. may increase operational cost) - High energy consumption for commixing process (i.e. solvothermal)	(Akram et al. 2022)
Pomegranate peel	Physically and chemically treated. Commixing with $LaCl_3 \cdot 7H_2O$ (10 mmol) and $FeCl_3 \cdot 6\ H_2O$ (5 mmol)	Phosphate (PO_4)	0.5 g/L	13.91 mg/g (Fe/Peel) 38.86 mg/g (La/Peel) 44.50 mg/g (Fe-La/Peel)	3 - 10	- The process is highly pH-dependent, which more effective at pH of 2.91 - The biosorbent has lower regeneration efficacy - Large chemicals consumption for chemical activation (i.e. may increase operational cost)	(Akram et al. 2021)

Materials	**Treatments**	**Type of pollutants removed**	**Adsorbent dosage**	**Max. adsorption capacity/percentage removal**	**pH**	**Drawbacks**	**Refs.**
Okara (soybean milk residues)	Physically and chemically treated.	Phosphate (PO_4)	1, 2, 3, 5, 7, 10, and 12 g/L	44.00 mg/g or 95% (dose 10 mg/L, 30 min, pH of 2 - 6)	2 - 12	- The process is more effective at acid pH (2 - 6) - At pH > 11 reduce the process efficacy due to strong competition of phosphate species and OH^- anions - Large chemicals consumption for chemical activation (i.e. may increase operational cost) - Potential environmental impact from chemical usage	(Nguyen et al. 2014)
Apple peel	Physically and chemically treated.	Phosphate (PO_4^{3-})	10 g/L	20.35 mg/g (or 100% at pH 2)	2 - 12	- The process is more effective at pH of 2 - Large chemicals consumption for chemical activation (i.e. may increase operational cost) - Potential environmental impact from chemical usage	(Mallampati and Valiyaveettil 2013)
Coconut shell fiber	Physically and chemically treated.	- Phosphate (PO_4^{3-}) - Sulfate (SO_4^{2-}) - Nitrate (NO_3^{-})	10 g/L	200 mg/g (phosphate) 31.2 mg/g (sulfate) 33.7 mg/g (nitrate)	5	- Large chemicals consumption for chemical activation (i.e. may increase operational cost) - The biosorbent has low adsorption constant and recyclability capacity than anionic resin	(de Lima et al. 2012)
Pesticide removal							
Delonix regia seeds	Physically and chemically treated. The resulted biosorbent was applied with P. stutzeri biomass (at ratio of 2:1)	Chlorpyrifos	25, 50, 75 and 100 mg/L	95.29% (in soil and water)	7.0	- Temperature > 30 ºC and pH > 7 may reduce the removal efficiency - The process is more complex to prepare mixed biosorbent - Large chemicals consumption for chemical activation (i.e. may increase operational cost) - Potential environmental impact from chemical usage	(Saravanan et al. 2022)
Potato peel	Physically and thermally treated (i.e. pyrolysis).	Chlorpyrifos	1, 2, 5, 10 g/L	46.02 (at 1 g/L)	5	- The process is highly pH-dependent (more effective at acid pH) - The efficacy of the biosorbent was gradually decreased in hemolysis in 24 h - High energy consumption for pyrolysis	(Singh et al. 2022)

Materials	Treatments	Type of pollutants removed	Adsorbent dosage	Max. adsorption capacity/percentage removal	pH	Drawbacks	Refs.
Maucaba waste (maucaba endocarp)	Physically (pyrolysis) and chemically treated.	Atrazine	0.1 g/50 mL	90 - 98%	-	- The making process of biosorbent is highly expensive - High energy consumption for pyrolysis - Large chemicals consumption for chemical activation (i.e. may increase operational cost) - Potential environmental impact from chemical usage	(Vieira et al. 2021)
Olive mill residue	Physically treated	- Imazalil (Im) - Thiabendazole (T)	0.1 g/100 mL	9 mg/g (Im); 8.6 mg/g (T)	7	- Type of biosorbent is highly influence the performance - High dose of adsorbent was used	(Fernández-López et al. 2021)
Artichoke waste				1.9 mg/g (I);6.6 mg/g (T)			
Citrus waste				6.6 mg/g (I); 6.1 mg/g (T)			
De-oiled Karanja seed cake	Physically and chemically treated.	2,4,6-Trichlorophenol	0.02, 0.03, and 0.04 g/L	74% or 124 mg/g	2, 6, and 10	- Higher temperature was found to reduce the process's efficacy - High pH (10) reduced the adsorption efficiency - The process was highly influence by pH, initial concentration, adsorbent loading, and adsorbent's particle size - Large chemicals consumption for chemical activation (i.e. may increase operational cost) - Potential environmental impact from chemical usage	(Aniya et al. 2021)
Eucalyptus bark	Physically treated	- Atrazine (A) - Imidacloprid (I)	30 mg/mL	83.80 – 88.80% (A) 70.40 – 75.40% (I)	-	- Type of biosorbent is highly influence the performance - High dose of adsorbent was used	(Mandal et al. 2017)
Corn cob				43.50 – 67.60% (A) 43.70 – 55.00% (I)	-		
Bamboo chips				48.00 – 58.70% (A) 53.80 – 87.20% (I)	-		
Rice straw				42.20 – 48.50% (A) 33.60 – 39.40% (I)	-		
Rice husk				42.20 – 57.70% (A) 50.70 – 58.80% (I)	-		
Apple shell				40.08 mg/g (for 2,4-DP) 17.86 mg/g (for 2,4-D) 25.64 mg/g (for 2,4-DB)	6.0		
Orange peel				22.71 mg/g (for 2,4-DP) 34.48 mg/g (for 2,4-D) 23.25 mg/g (for 2,4-DB)	6.0		

Materials	Treatments	Type of pollutants removed	Adsorbent dosage	Max. adsorption capacity/percentage removal	pH	Drawbacks	Refs.
Banana peel				33.26 mg/g (for 2,4-DP) 22.73 mg/g (for 2,4-D) 21.27 mg/g (for 2,4-DB)	7.0		
Pharmaceutical compounds removal							
Tomato waste (TW)	Physically treated	- Praziquantel (PRAZ) - Febantel (FEBA) - Procaine (PROC) - Dexamethasone (DEXA) - Tylosin tartrate (TYL)	100 g/L	449 µg/g (PRAZ) 524 µg/g (FEBA) 1230 µg/g (PROC) 461 µg/g (DEXA) 677 µg/g (TYL)	4.58	- Longer contact time is needed (at 24 hrs contact time) - High dose of biosorbent was used - Reduced performance efficacy due to the ’salting out’ effect of the surface of TW biosorbent	(Mutavdžić Pavlović et al. 2021)
Maple leaves (ML)	Physically treated	Ciprofloxacin	0.5 – 1.5 g/20 mL (with 0.5 g increment)	519.84 – 2773.10 mg/g	3-11	- The process was highly influence by pH - Optimum performance at pH of 3 and 11 - The lowest adsorption capacity from TP was due to its smallest surface area	(Tolić et al. 2021)
Tomato waste (seeds and peel)				60.57 – 97.05 mg/g			
Tangerine peel (TP)				30.21 – 77.08 mg/g			
Coconut husk	Physically and chemically treated.	2-(4- Isobutyl phenyl) propanoic acid (or known as Ibuprofen)	1 g/L	76.92 mg/g	-	- Large chemicals consumption for chemical activation (i.e. may increase operational cost) - Longer contact time is needed (at 60 minutes contact time)	(Bello et al. 2020)
Peanut shell biochar (PSB)	Physically and thermally treated (i.e. pyrolysis alone (800-PSB) or combined with hydrothermal carbonization (190-800-PSB and 800-800-PSB))	Naproxen	0.5 g/L	- 105 mg/g at pH 8.91 (800-PSB) - 215 mg/g at pH 8.80 (190-800-PSB) - 324 mg/g at pH 9.15 (800-800-PSB)	3 - 11	- Optimum performance under basic pH - Large chemicals consumption for chemical activation (i.e. may increase operational cost) - The potential of micropore blocking due to naproxen adsorption - High energy consumption	(Tomul et al. 2020)
Soybean husk	Physically and chemically treated.	Ibuprofen	20 g/L	50 mg/g	4.75	- High dose of adsorbent was used - Large chemicals consumption for chemical activation (i.e. may increase operational cost)	(Bello et al. 2019b)
Banana peel	Physically treated	- Salicylic acid (SA) - Benzoic acid (BA)	1.25 - 4.00 g/50 mL (with 0.25 -0.50 g increment)	61.55 % (SA) 77.59% (BA)	2 - 11	- Longer contact time is needed (at 720 and 840 minutes for BA and SA) - Optimum performance at pH of 3 - The process efficacy reduced at pH > 3 - High dose of biosorbent was used - Both biosorbent has low adsorption capacity	(Pathak et al. 2016)

Materials	Treatments	Type of pollutants removed	Adsorbent dosage	Max. adsorption capacity/percentage removal	pH	Drawbacks	Refs.
Rice straw (RS)	Direct use of rice straw and gypsum, then mixed with top soil. Leachate was used as wastewater	- Electrical Conductivity (EC) - Cations (i.e. Ca^{2+}, Mg^{2+}, K^{+}, and Na^{+})	n.a.	Improved salt-leaching efficiency: - EC was reduced by 80.19% - Ca^{2+} reduced by 69.85% - Mg^{2+} reduced by 81.03% - K^{+} reduced by 22.07% - Na^{+} reduced by 86.48%	7.80 - 8.95	- Use of RS alone did not have higher efficacy than mixed RS+G - Addition of gypsum improved performance, but may increase chemicals consumption hence the operation cost	(Ebrahim Yahya et al. 2022)
Rice straw with gypsum (RS+G)			1.4 RS added with 0.005 kg G	Improved salt-leaching efficiency: - EC was reduced by 92.01% - Ca^{2+} increased by 519.12% - Mg^{2+} reduced by 75.26% - K^{+} reduced by 24.14% - Na^{+} reduced by 96.47%			
Coconut shell (CS)	Physically treated	Electrical conductivity	CS adsorbent was placed in the column (Ø=10.10 cm) with 50 cm thickness	After 2 hours, EC reduced from: - 6,132 µS/cm to ~1500 µS/cm (100% urine sol.) - 4,682 µS/cm to ~1750 µS/cm (50% urine sol.) - 4,035 µS/cm to ~2000 µS/cm (20% urine sol.)	8-9.5	- After 16 hrs contact time, EC increased to the initial value - CS biosorbent was not highly efficient because the effective adsorption of dissolved salt (i.e. EC) only occurred at 2 hrs of contact time	(Nguyen et al. 2021)
Wheat straw	Physically treated	Sodium	0.3, 0.5, 0.7, 1.0, 1.3, and 1.6 g/40 mL	90.43% (at dose > 0.5 g/40 mL)	3-8	- Best performance at pH 5 - Adsorption rate was almost constant after 30 minutes contact time - Increasing initial concentration of sodium solution reduced adsorption capacity	(Rasouli et al. 2020)
Rice husk				90.37% (at dose > 0.5 g/40 mL)			
Pine wood (PW)	Physically and chemically treated. Modification of hemicellulose (HC) extraction using DTPA, and chitosan (CS) was added to improve saline adsorption.	NaCl (0.3% and 0.9%)	0.4 g/L	HC−DTPA− CS, PW: - 19 g/g (0.3% NaCl) - 27 g/g (0.9% NaCl)	3.9	- Increasing temperature increases the NaCI adsorption - Large chemicals consumption for improving performance efficacy	(Ayoub et al. 2013)
Switch grass (SG)				HC−DTPA, SG: - 3 g/g (0.3% NaCl) - 5 g/g (0.9% NaCl) HC−DTPA− CS, SG: - 23 g/g (0.3% NaCl) - 27 g/g (0.9% NaCl)			
Coastal bermuda grass (SBG)				HC−DTPA− CS, CBG: - 23 g/g (0.3% NaCl) - 28 g/g (0.9% NaCl)			

Heavy metals

The biosorption process for heavy metal removal followed some of the mechanisms, for instance, ion exchange, adsorption, complexation, chelation, and precipitation (Agarwal et al. 2020). The mechanisms present in the heavy metal biosorption process depend on the functional groups of biosorbent. This functional group attracts metal ions towards its surface. The functional groups found in agricultural waste biosorbent are hydroxyl, carboxyl, amide, amine, imine, phosphates, hydroxyl, carbonyl, sulfhydryl, and phenolic group (Rao and Khan 2009; Yuvaraja et al. 2014). The biosorption process of heavy metals always includes interaction between two or more mechanisms. It is quite rare to find just one particular mechanism because of the complex chemistry involved. The heavy metal removal efficiency was affected by temperature, pH, contact time, sorbent dose, initial metal concentration, and stirring rate (Beni and Esmaeili 2020; Rao and Khan 2009; Song et al. 2017). Heavy metals biosorption followed two general steps. The first step is the movement of heavy metals in solution caused by physical attraction of heavy metals to the surface of a biosorbent that has a negative charge. The second step is the transfer, followed by the bonding between the dissolved component from the biosorbent surface and the active sites (Beni and Esmaeili 2020; Noli et al. 2019).

Ion-exchange

Ion exchange is reported as the main mechanism involved in heavy metal biosorption. An ion-exchange reaction occurs between ions on the surface of the biosorbent with heavy metal ions in the solution. Metal ions such as Cu(II), Pb(II), Cd(II), Ni(II), Dy(III), Zn(II), Fe(III), Cr(III), Mn (II), Co(II), Hg(II), Ag(I), Th(IV), U(II) and U(VI) can be removed from wastewater through this mechanisms. The general reaction of the ion-exchange mechanism of heavy metal is illustrated by the equation (Agarwal et al. 2020; Singh et al. 2020):

$$M^{X+} + X(CD) \leftrightarrow XC^{+} + MD_{X}$$

Where M^{X+} = heavy metals ion, MD_X = sorbed M^{X+}, and CD = a number of acid sites on the surface of biosorbent (Agarwal et al. 2020; Zhou et al. 2016).

In a study of heavy metal biosorption through ion-exchange mechanisms, the residue from brown seaweed alginate extraction has been used for cadmium removal. Light metals such as Ca, Mg, Na, and K are present in the biosorbent surface. The ion-exchange mechanism is linked with the carboxyl group involving mainly Ca ions since Ca content decreases while Cd ions appear after biosorbent contact with Cd solution. These reactions were also proved with the deposition of calcium compounds. Though ion exchange is the main mechanism, it also includes other mechanisms such as complexation and chelation (Nishikawa et al. 2020). Another study that removed Copper using Macrocystis pyrifera as biosorbent revealed an ion-exchange mechanism between Ca and Mg with Cu(II). As a primary mechanism, ion exchange accounted for 56% of the total Cu(II) bound to the surface of the biosorbent (Cid et al. 2020).

Adsorption

Adsorption is not a predominant mechanism found in biosorption compared with other mechanisms. The adsorption mechanism can be distinguished into two different processes based on the forces, physical adsorption, and chemical adsorption. Heavy metals ions such as Cu(II), Zn(II), Cr(III), Cr(IV), Cd(II), Pb(I), Pb(II), Dy(II), Fe(III), Ag(I), Ni(II) are reported undergo physical adsorption mechanism. A reversible reaction via van der Waals forces mediates the weak binding of biosorbent surface with heavy metals. However, some studies reported this adsorption as a primary mechanism for their heavy metal removal. An example was reported for Cu(II) removal from wastewater using rubber leaf powder as a biosorbent. The adsorption mechanisms favor low temperature since high temperatures cause Cu (II) ions to escape from the biosorbent surface to the solutions. Other factors such as pH, initial metal concentration, biosorbent dosage, and particle size also affect adsorption (Wan Ngah and Hanafiah 2008). From the thermodynamic point of view, this adsorptions mechanism is an exothermic reaction with a negative value of enthalpy change and spontaneous reaction since it has negative Gibb's free energy (Al-Anber and Matouq 2008). The chemical adsorption mechanism is a non-spontaneous and endothermic reaction, unlike physical adsorption. The chemical adsorption was a dominant mechanism in removing Ni(II) using Litchi chinensis seeds as biosorbent. Ni(II) formed a strong chemical bond with the active sites on the biosorbent surface. This reaction needs an input of energy for the forming chemical bond to take place (Flores-Garnica et al. 2013). Other heavy metals reported removed by chemical adsorptions are Cr(IV), Pb(II), Cd(II), and Zn(II) (Amar et al. 2020; Oliveira et al. 2021).

Surface precipitation

The surface precipitation mechanism generally occurs when the heavy metals concentration exceeds the biosorbent capacity. Insoluble heavy metal hydroxides and oxides formed on the surface of the biosorbent as precipitation took place. The accumulation of heavy metals will be possible when there is a negative charge on the surface of the biosorbent (Schneider et al. 2001). Removal of some heavy metals such as Cr(III), Cu(II), Cd (II), U(VI), La(III), Sm(III) were reported to implicate surface precipitation as one of the biosorption mechanisms. A study using soybean meal waste as biosorbent revealed that surface precipitation takes place when the Cr(III) and Cu(II) ion concentration increases exceeding the capacity of binding the functional groups in the biosorbent surface. The kineticity of this precipitation reaction is much slower compared with other mechanisms (Witek-Krowiak and Reddy 2013). Another study using Aloe vera wastes treating U(VI) and Cd(II) reported the occurrence of the surface precipitations mechanism along with ion exchange and physical sorption facilitated by carboxyl, carbonyl, and hydroxyl functional groups (Noli et al. 2019).

Complexation

The heavy metal complexes consist of metal ions as a center bound to one or more ligands. The formation of a heavy metals complex in the biosorption process requires electron sharing between heavy metals and functional groups of biosorbent as ligands. The central metal coordinated by ligands can be one single metal (mononuclear) or more (polynuclear). These mechanisms reported playing a role in several heavy metals removal (Cr(IV), Pb(II), Zn(II), Cu(II), Ag(I), Tb(III), Th (IV), La(III), Sm(III)) though not necessarily as the main mechanism (Krishnani et al. 2008; Lu and Guo 2019; Noli et al. 2019). Biosorbent derived from rice husk contain lignin and cellulose in the biomatrix bearing carboxyl, alcohol, and ketones functional groups. These functional groups act as ligands and form complexes with Ni(II) and Cu(II) (Krishnani et al. 2008). Another study used the walnut shell to treat Pb(II). A modification of walnut shell using xanthate led to the higher adsorption performance caused by better complexation and ion exchange. Formation of the complex increases because of xanthogenization of walnut shell grafted sulphur-containing functional groups on the walnut shell surface. Based on the XPS and FTIR analysis, the sulfhydryl group (-SH) reacts as ligands and makes a complex with lead ions (Lu and Guo 2019).

Chelation

Chelation is a binding reaction of organic molecules (ligands) and metals ions to form a complex ring-like structure. As a chelating agent, the ligand formed a covalent and coordination linkage with the metal's ion. Several atoms like S, N, and O are commonly found to be an active site of the functional groups, such as RCOO-, -SH, S-S, OH, P(=O) $(OR)_3$, and NH_3 (Flora and Pachauri 2010). Among several functional groups, NH_3 has the highest effectiveness for heavy metals removal, caused by the combination of the chelation process with electrostatic interaction and hydrogen bonding (Arief et al. 2008).

Chelation's mechanisms are affected by both metal's ion and chelating agent's properties. Occupation of more of the coordination points of metals ion increases the stability of the complex formed. Metals ions like Cr(III), Cr(IV), Cu(II), Pb(II), Cd(II) form a stable structure with chelation agents by the formation of multiple bonding (Bulgariu and Bulgariu 2018; Mata et al. 2009). A recent study employs a chelation mechanism to remove heavy metals using soy waste biomass. Industrial sulphur chelating agents were used for biosorbent functionalization. The functionalization increases the number and availability of functional groups responsible for the metal's recovery. FTIR analysis showed that functionalization causes spatial reorientation of some functional groups like carboxyl, carbonyl, hydroxyl, and amino. Moreover, functionalization is also causing new sulphur-containing functional groups (mainly Thiol groups) to appear. The performance of heavy metal removal for Pb(II), Cu(II), and Ni(II) has doubled after functionalization due to the increased affinity of biosorbent surface to heavy metals (Bulgariu and Bulgariu 2018). Chelation mechanism also reported in pectin gels biosorbent derived from the sugar-beet pulp. The polymeric structure of the pectin gel is rich in a carboxyl group that acts as a chelating agent for Pb(II). FTIR spectra of pectin gel showed that the two carboxyl band distance decreased after being treated with led ions, indicating the chelation reaction occurrence (Mata et al. 2009).

Dye

Jawad et al. (2019) studied the mechanism of biosorbent made of carbonized watermelon rind to remove methylene blue (MB) dyes in an aqueous solution. Their study found that there were three interactions of the MB adsorption mechanism, including:

1) electrostatic attractions between negatively charged functional groups on the biosorbent surface area with positively charged MB cations;
2) hydrogen bonding interaction between the surface hydrogen bonds available on biosorbent surface and the nitrogen atoms available on the MB; and
3) π–π stacking interactions between aromatic rings of MB and the graphene framework of biosorbent.

The different interaction routes influence the efficacy of MB dyes adsorption onto the surface of the biosorbent. Such a mechanism was also reported in other studies (Fan et al. 2017; Jia et al. 2016; Liu et al. 2018; Üner et al. 2016). Liu et al. (2018) described that the adsorption mechanism of MB dyes molecules onto activated carbon from tea waste was rather complex, involving a fast- and slow-stage. The fast-stage is composed of tea waste's electrostatic ion exchange between MB and organic functional groups. At this stage, the adsorption can be achieved within 5 minutes contact time with the MB removal rate of 79%. At a slow-stage, which can be finished by the remaining time with the MB removal rate of 21%, was mainly due to the hydrogen bond or π-π stacking interaction between MB and tea waste. This study suggested that the electrostatic ion exchange was not the only mechanism in removing MB dyes. However, the organic functional groups derived from tea waste also played a critical part in MB adsorption onto tea waste. Another study by Ardekani et al. (2017) reported that the ultrasound-assisted removal method combined with activated carbon wood of cherry trees resulted in a significant increase in the efficiency of the MB removal. Their study observed that an increase in pH and contact time was responsible for decreasing the positive charge on the surface and increasing the number of negatively charged sites. Such conditions contributed to enhancing the MB removal mainly due to the electrostatic ion exchange interactions.

Pesticide

Bezerra et al. (2020) explained that removing herbicide using Moringa oleifera seed husks biosorbent was due to an instant adsorption process through the biosorbent's boundary layer. However, the pore diffusion step may hinder the process. The study also found that an increase in operating temperatures increases the removal efficiency. Similarly, the biosorption mechanism of pesticide using ACR or AIW biosorbent involved ion exchange, chelation, and complexation with the functional groups on the biosorbent's surface and the release of H_3O^+ into solution (Sarker et al. 2017). Various factors influence biosorption efficacy, including biosorbent dosage, initial pollutant concentration, pH, temperature, contact time, biosorbent particle size, etc. (Bezerra et al. 2020; Ramrakhiani et al. 2019; Sarker et al. 2017). According to Ramrakhiani et al. (2019), the glyphosate herbicide removal using biosorbent was possibly due to the formation of phosphate-metal complexes as (1) mononuclear or monodentate (bridging); (2) binuclear or bidentate surface complexation; or (3) dense-packed mononuclear surface complexation at increasing concentration of glyphosate herbicide. Another route is the formation of metal-carboxyl group and surface-amino group complexes, which can also be considered as weak binding. Chen et al. (2019) demonstrated that biosorbent made of peanut shell or wheat straw immobilized laccase could remove nine pesticide residues (i.e. isoproturon, atrazine, prometryn, mefenacet, penoxsulam, nitenpyram, prochloraz, pyrazosulfuron-ethyl, and bensulfuron-methyl). They further added that the pesticide removal mechanism involves biosorption mechanism which coupled with degradation.

Organic pollutants

Liu et al. (2017) studied the application of biochar from poplar catkin as a low-cost and renewable biosorbent in removing organic pollutants such as organic compounds from wastewater streams. The study demonstrated that the adsorption of organic pollutants was endothermic and spontaneous, which comprised outer-sphere surface complexes and hydrogen-bonding interactions.The findings confirmed that converting poplar catkins into biochar can be a sustainable solution in removing organic pollutants in wastewater. While a study by Dawood et al. (2017) found that the adsorption mechanism of organic pollutants removal using pine cone biochar was mainly following the chemisorption route, endothermic and spontaneous. Such adsorption mechanisms of organic pollutants have also been highlighted in various studies (Chen et al. 2019; Dai et al. 2019; Elayadi et al. 2020).

Opportunities and challenges for future implementation

Wastewater is widely used in the world for irrigation due to the water scarcity and the demand for the wastewater treatment is increasing rapidly (Tabatabaei et al. 2020). ACR and AIW are highly abundant in terms of availability in Indonesia and can be used for the treatments. Most ACR and AIW are the carbonaceous source that is potential for activated carbon, for instance, coconut shells (Grace et al. 2016). Many studies have reported that ACR or AIW, either as filter media, activated carbon, biochar, or biosorbent, are effective for removing heavy metals, organic and non-organic pollutants, and dyes (Rodriguez et al. 2020; Sadeek et al. 2015; Yahya et al. 2015; Yap et al. 2017). Grace et al. (2016) and Bhatnagar et al. (2010) reviewed, for instance, that coconut shells were widely used for removing contaminants (i.e. nitrate, ammonium, and phosphate) in drinking water treatment or pollutants (i.e. Cu, Pb, Cd, Zn, Ni, dyes, natural organic matter/NOM, radionuclides, and anions) in wastewater treatment. Another successful example of valorizing of AIW was tea and coffee waste activated carbon which effectively removed phenol and NO_3^--N in wastewater streams (Lamine et al. 2014; Wang et al. 2014). Various studies, for instance, also highlighted the potency of ACR such as rice husk as a successful biosorbent for removing nitrate, phosphate, ammonium, heavy metals, dyes, and humic acid (Daifullah et al. 2004; Grace et al. 2016; Krishnani et al. 2008; Liu et al. 2012). Yahya et al. (2015) reported that ACR is relatively inexpensive, locally available, and effective materials to be commercially used as activated carbon for many applications, including wastewater treatment. Bolong et al. (2016) stated that using OPEFBs as biosorbent for simple wastewater treatment technology offered a sustainable solution for waste problems faced by oil palm mills and offered alternative eco-friendly and high value-added bioproducts. Those studies revealed a great potential of using ACR or AIW in water or wastewater treatment.

However, the critical challenges of transforming ACR and AIW in wastewater biosorption technology include the high cost of carbonation and activation process, making the process not sustainable and feasible. Also, the need for regeneration treatment as the biosorbent tends to become saturated and exhausted, which reduces the efficacy of pollutant removal (Grace et al. 2016; Rashid et al. 2016). Another challenge is that further investigation is required on the production, optimization, and applications of ACR- and AIW-based wastewater biosorption on a much bigger scale and to prove their valuable application (Yahya et al. 2015). Rashid et al. (2016) stated that the difficulty of biosorbent to recover during and at the end of analysis restricts its commercial application.

Conclusion

The application of ACR and AIW as natural biosorbent, filter media, activated carbon, or biochar in wastewater technology is greatly potential. Indonesia has abundant ACR and AIW, which are currently and mainly disposed of to landfills or surrounding areas. Therefore, the availability, renewable resources, and low-cost raw material acquisition of ACR and AIW pose huge opportunities for further valorisation into more high-added value products. Yet, further investigation is essential to examine the scaling-up and commercialization potential of ACR and AIW as natural biosorbent in wastewater treatment. This paper supports that valorizing ACR and AIW for treating wastewater provides multiple benefits of reducing waste problems, implementing eco-friendly and sustainable wastewater treatments, and providing social and health benefits to nearby society. Thus, the broader application prospect of low-carbon and low-cost ACR and AIW utilization in wastewater biosorption is greatly feasible.

Compliance with ethical standards

Conflict of interest The authors declare that there are no conflicts of interest associated with this study.

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