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Pyrolyzed Biochar Nanocomposites as Pollutant Adsorbent: A Review

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Abstract— The emergence of new pollutants such as pharmaceutically active waste and increasing antimicrobial resistance in bacteria are most recent environmental issues, The persistent nature and mode of action of pharmaceutically active compounds in the environment make them a serious concern. The Heavy volumetric production of Drugs by pharmaceutical industries and subsequentially it's widespread utilization in health care centres, hospitals, diagnostic centres and veterinary hospitals, among others, have significantly contributing the part in augmentation of pharmaceutical active residues in environmental. Pharmaceutically active compounds (PACs) are released into the environment via various pathways; wastes generated by humans and animals are the major sources. Herein the category of some pharmaceuticals and its adverse effect because of free exposure in the environment included. Various type of techniques is being used for wastewater treatment, among of them the adsorption technique is more effective and easier to operate than others. The commercial activated carbons are economically expensive, which is the limiting factor for use of adsorption technique; hence the exploration of sustainable materials that are cost-effective, ecofriendly, efficient, easy to use and socially acceptable for real-scale environmental applications is currently required. Biochar made up from agricultural wastes offer such advantages as abundance, sustainable, low-cost, and eco-friendly materials. Currently, Biochar as one of the main pyrogenic products from thermo-chemical process of ligno-cellulosic biomass has been widely studied to deal with water pollutants and wastewaters. Physically or chemically modified biochar composites can effectively increase the yield of antibiotics adsorption. Furthermore, it could be regenerated and reused by efficient management, which is the significant advantage of biochar-based adsorbents. This review, therefore, attempts to provide an overview of the current interests in the applications of biochar for the removal of PACs like antibiotics from wastewater.

Keywords—Pharmaceutical waste, Antibiotics, Adsorption, Biochar, Pyrolysis.

I. INTRODUCTION

This Through the years, lives have been saved thanks to the discovery of antibiotics and their application in treating diseases, notably bacterial infections.[1] A huge variety of drugs are now being used by humans daily. The Pharmaceutical Industry of India is 3rd largest in terms of the volume production in world. India is one of the largest exporter of generic drugs in volume terms, is also the third-largest API manufacturer, behind China and Italy. According to the pharmexcil (pharmaceutical export promotion council) there is an estimation that one out of five drugs have an Indian connection which are globally sold out. Higher amount of treated or untreated waste water disposed by these manufacturing industries that contains various types of pollutants. The effluent from these pharmaceutical industries, hospitals, health care centres, laboratories is raising day by day.

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Waste water which is released from pharmaceutical industries, hospitals and health care centres is becoming an emerging issue in this present era. Waste water from them contains degradable and nondegradable pollutant components. These effluents are released in environment without treatment and it contains a huge variety of harmful chemical substances which can cause risks to living things or environment. The release of pharmaceutical waste water directly or untreated, contaminates surface water and also mix with ground water. Recently, a preliminary list of emerging substances, containing the classes of pollutants are; antioxidants, anticorrosive, nanoparticles, industrial chemicals, food additives, flame retardants, detergents antifoaming agents, antifouling compounds, gasoline additives biocides, bio terrorism/sabotage agents, complexing agents, , disinfection by-products (drinking water), drugs, fragrances, , perfluoro alkylated substances and their transformation products, pesticides, plasticizers, personal care products, trace metals, pharmaceuticals, and their compounds, wood and food preservatives, among others [2]. Among all pollutants in water, pertinent attention has been made to pharmaceutical effluents. The increasing informative broad spectrum of product and toxicity [3,4] due to their unpredictable impact on environment, even when they are present at very low concentration-levels.

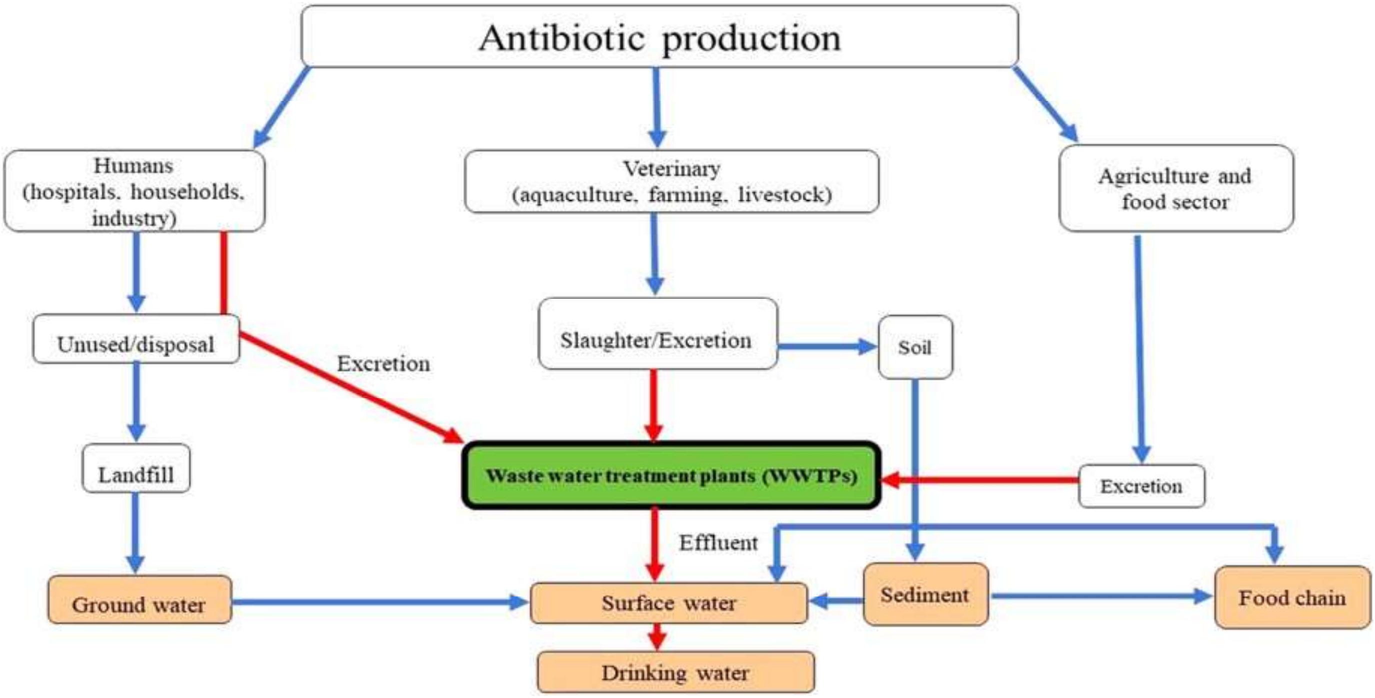

Fig. 1. Routes and sources of antibiotics in the environment (Adapted with permission from Ref. (Harrower et al., 2021))

Worldwide population is seriously affected from the untreated or partially treated disposal of waste water in direct and indirect ways. People do not have proper facilities to decontaminate the polluted water sources which cause serious threat to their health. People utilize contaminated water for their regular needs. In the developing countries, the people are not able to continuously access fresh water sources which leads to the increasing diseases related to water. As indicated by WHO, around 1.6 million people are dying year after year because of the various diseases caused by polluted water and the 90% of the children under 5 are more susceptible to this disease (Pandit & Kumar 2015; WHO 2017)

Here we focusing on the pollution which is created by pharmaceutical waste or drugs residues in discharged water. In recent years, several articles have been reported regarding the drugs/pharmaceuticals in the environment [5]. Drugs are considered as the most non-biodegradable chemical compounds in the environment [6]. Most of the pharmaceuticals, being insoluble in water, are not effectively washed away by rain. Therefore, drug residues remain in the environment and eventually accumulate in the bodies of terrestrial and aquatic organisms [7]. Several classes of drugs such as histamine blockers, di-uretic, antibiotics, anti-inflammatory drugs, antifungal drugs, sulpha drugs, antidiabetics, anti-allergic drugs, barbiturates, β-blockers, hormones, antihypertensive drugs, lipid regulators, histamine blockers, psychiatric drugs, topical products and antiseptics have been detected and monitered in surface waters [8–25]. Some drugs/pharmaceuticals have also been reported in drinking waters [26-28]. Although almost all of the pharmaceuticals are normally detected at low concentrations but their continuous discharge into the surroundings may

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increase the possibility of their synergistic effects with other pharmaceuticals or chemicals in the aquatic environment [29]. As the population raising exponentially same as use of drugs also raises in same way. Among the other drugs antibiotics are more popular drugs for their amazingly effective anti-infective works. Since antibiotics are not completely digested after consumption, they can be discovered in sewage systems [30]. These non-digested antibiotics are frequently not biodegradable and photolysis is useless against them. According to a recent study, India has been the world's most substantial end-user since 2015, posing a serious threat to their resistance [31-35]. Antibiotics can be detected in water samples due to ineffective conventional methods of elimination from wastewater [36-37]. However, some metabolized antibiotics are eliminated through defecation while the active nonbiodegradable residues accumulate greatly, developing bacteria with antibiotic-resistance [38].

As a result, antibiotic resistant germs kill thousands of people yearly worldwide. Antimicrobial resistance reported in detectable concentrations in drinkable water is lower than that found in numerous effluents, such as hospital discharges,

[32,37,39]. Health center wastewater consists of composite mixture of various toxic compounds that include antihistamines, hormones, immunosuppressive drugs, pharmaceuticals and their metabolites, cytotoxic agents, disinfectants, antibiotics releasing from diagnostic, autopsy centers and research activities and generally in the form of medical excreta from patients (Verlicchi *et al*., 2010b; Ortolan, 1999).

Several conventional water techniques have been employed for the elimination of pollutants and are reported to be accompanied by many drawbacks (Inyinbor et al., 2022; Moradi and Sharma, 2021; Rajabi et al., 2019). These techniques include chemical coagulation accompanied by the formation of sludge with high content of salts used as coagulants (Kooijman et al., 2020; Qian et al., 2020), ion exchange disadvantaged by high operational costs, advanced oxidation process which requires a large quantity of reagents for its operation (Wang, 2016), reverse osmosis is known to be expensive (Anis et al., 2019), electrocoagulation produces sludge (Zaied et al., 2020), constructed wetland process requires a large area of land (de Oliveira et al., 2019; Lancheros et al., 2019; Yan et al., 2018) and biological treatment process slowly progresses (Bhatia et al., 2018; Gholizadeh et al., 2020).

In contrast, adsorption processes are inexpensive and efficient (Dada et al., 2013; Inyinbor et al., 2019a, 2019b, 2019c) and have become the most preferred technique due to their ease of operation, adaptability and simple design (Anijiofor et al., 2018). Also, developing cheap and effective adsorbents will make adsorption globally accessible. Hence, a huge quest for natural, readily available, eco-friendly and low-cost materials as adsorbents (Bai et al., 2021; Ronda et al., 2015; Wang and Chen 2014; Xu and Wang 2017). Agricultural wastes are of low or no economic importance (Adeniyi et al., 2020; Adeniyi and Ighalo 2019), and liters the environment. Therefore, their clean-up is necessary. Agricultural wastes are lignocellulosic and carbonaceous in nature with abundant surface functional groups, which qualifies them as alternatives to commercial activated carbon (Inyinbor et al., 2019a, 2019b, 2019c). Many adsorbents have been produced from different waste entities such as rice husks (Li and Xiao 2019; Ng et al., 2019; Reddy et al., 2017), avocado peels (Palma et al., 2016; Salomo´n-Negrete et al., 2018), orange peels (Ahmed et al., 2020), neem husk (Mandal et al., 2020; Marichelvam and Azhagurajan 2018), cow faeces (Kaur et al., 2016; Mohd Nasir et al., 2019), tamarind fruit shell (Ashok et al., 2020), cotton seed hull (Yahya et al., 2020), banana fronds (Ali 2017; Ali et al., 2016) and many others (Anijiofor et al., 2018).

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Table 1: Different type of antibiotics and their adverse effect

Pharmaceutical active compounds	**Therapeutic use**	**Adverse effect in environment**	**References**
Ciprofloxacin (CFX)	Synthetic human antibiotic	Negatively affect the surface water and groundwater quality Nucleic acid synthesis inhibition	[40,41]
Sulfamethoxazole (SMX)	Human and veterinary antibiotic	Bioaccumulation in aquatic organisms Induces antibiotic resistant genes in various organisms Folic acid metabolism blockage	[42,43]
Chlortetracycline (CTC)	Veterinary antibiotic	Adverse effect on various aquatic organisms like histological alteration in gills of fish by pro-oxidative activity development of antibiotic resistance in various bacteria	[44]
Levofloxacin (LEV)	Bacterial antibiotic	Antibiotic resistance in humans and animals	[45,46]
Amoxillin (AMX)	Human and veterinary antibiotics	Cell wall synthesis inhibition in organism as concerning side effect	
Erythromycin (ERY)	Human antibiotic	Protein synthesis inhibition and metabolism disfunction in organisms	

These waste products are released from, industrial, domestic and / or agricultural operations. This review, therefore aims at summarizing the occurrence, the impacts of pharmaceuticals and the exploration of different agricultural wastes for the adsorptive removal of different pharmaceutical residues from wastewater.Hence, this review tried to establish a look ahead into preserving the environment from the threat of emerging contaminants such as pharmaceutically active compounds were included below in Table 1

II. BIOCHAR

In recent years, to control antibiotics concentration level of effluent waste water the scientific community has led to develop their significant interest in sustainable, non-toxic, inexpensive and easily available agriculture waste biomass material. Agricultural biomass works efficiently for the removal of residual antibiotics from waste water via adsorption process. [47] Agriculture biomass waste is the waste residual parts obtained from forests, landfills or fields.

Adsorbents produced from biomass possess functional sites that expedite the removal of pharmaceutically active compounds present in waste water . Biochar is a pyrolyzed carbon rich product of agriculture waste biomass in an inert atmosphere. Over the recent years interest is growing Fastly towards biochar for its intrinsic properties such as porous structure, high specific surface area, surface functionality and huge possibility of modifications.[48] biochar excellently performs the adsorption of aqueous pollutants. All of its properties are mainly depending on raw biomass matter type, catalysts, temperature, carrier gases.[49] Due to advances in research, miscellaneous methodologies have been developed to determine the properties of biochar depending on the method of preparation and modification. From that they can inaugurate better adsorption performance. [49,50]

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ADSORPTION PROCESS:

The adsorption process of contaminant molecules onto agriculture biomass-based adsorbent has follows subsequent principal steps, in which: Adsorbate molecule moves from matrix. Adsorbate molecule diffuses through the liquid membrane surrounding the adsorbent. Adsorbate molecule moves to the active sites of adsorbent Adsorbent-adsorbent interaction While the mechanism of adsorption depends on the latter step of whole mechanism. [49,51] Different type of interactions is responsible for antibiotics adhering to the surface of biochar examples like- p-p electron-donor accepter interaction, electrostatic, charge-dipole interaction, hydrophobic interactions. Some other factors like functionality, process conditions, temperature, pH, nature of contaminants, pore structure, competing ions, are also postulated to be responsible for the fluctuation in mechanism and adsorption capacity of adsorbent. [52,53,54] It is therefore, essential to choose more efficient adsorbent for adsorption of target antibiotics. [55,56] The interactions involved in the sorption process of adsorbate molecules onto biochar material surface are illustrated in following fig. 1

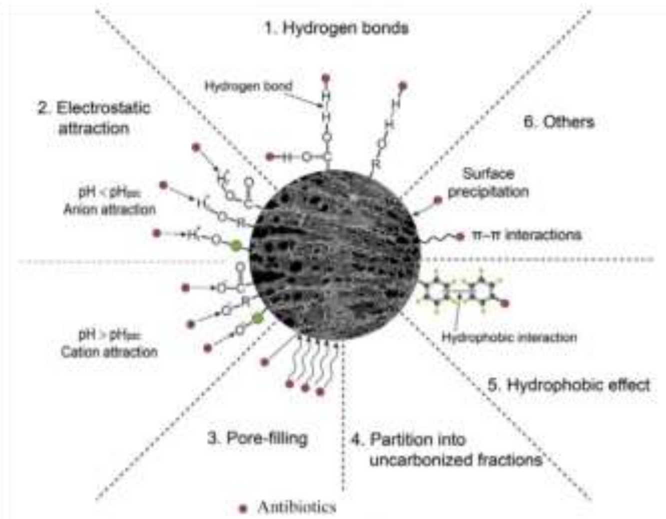

Fig. 2. Possible mechanisms during the adsorption of antibiotics (Adapted with permission from Ref. (Tan et al., 2015))

BIOCHAR AS ADSORBENT:

There are different methods to improve the structure and efficiency of biochar-based adsorbents. Based on the modifications and mechanisms of the synthesis, the obtained substances could be classified as pure/original BCs and modified or treated BCs, which allows the second group to be divided into modified biochar and biochar composites (Krasucka et al., 2021).

i. Pure Biochar

Pure biochar is made through slow pyrolysis of biomass under a maximum temperature of 700 °C, in the absence or presence of oxygen (less than 2%) (Ahmed et al., 2015; Major et al., 2009; Manyà, 2012) via two methods- physical methods (microwave pyrolysis, grinding and ball milling) or chemical methods using reductants NaOH, KOH or oxidants such as HCl, H_3PO_4, H_2SO_4, or during pre- or post-synthesis. Some pristine biochar often associated with lack affinity for antibiotic pollutants, because of their low porosity as an adsorbent and absence of functionally sufficient surface area, so they are referred to modifications.

ii. Modified Biochar

Pure BC-based adsorbents showed an unacceptable capacity for the adsorption of different antibiotics. There is lack of interaction between the adsorbent and medicine related issue occur.

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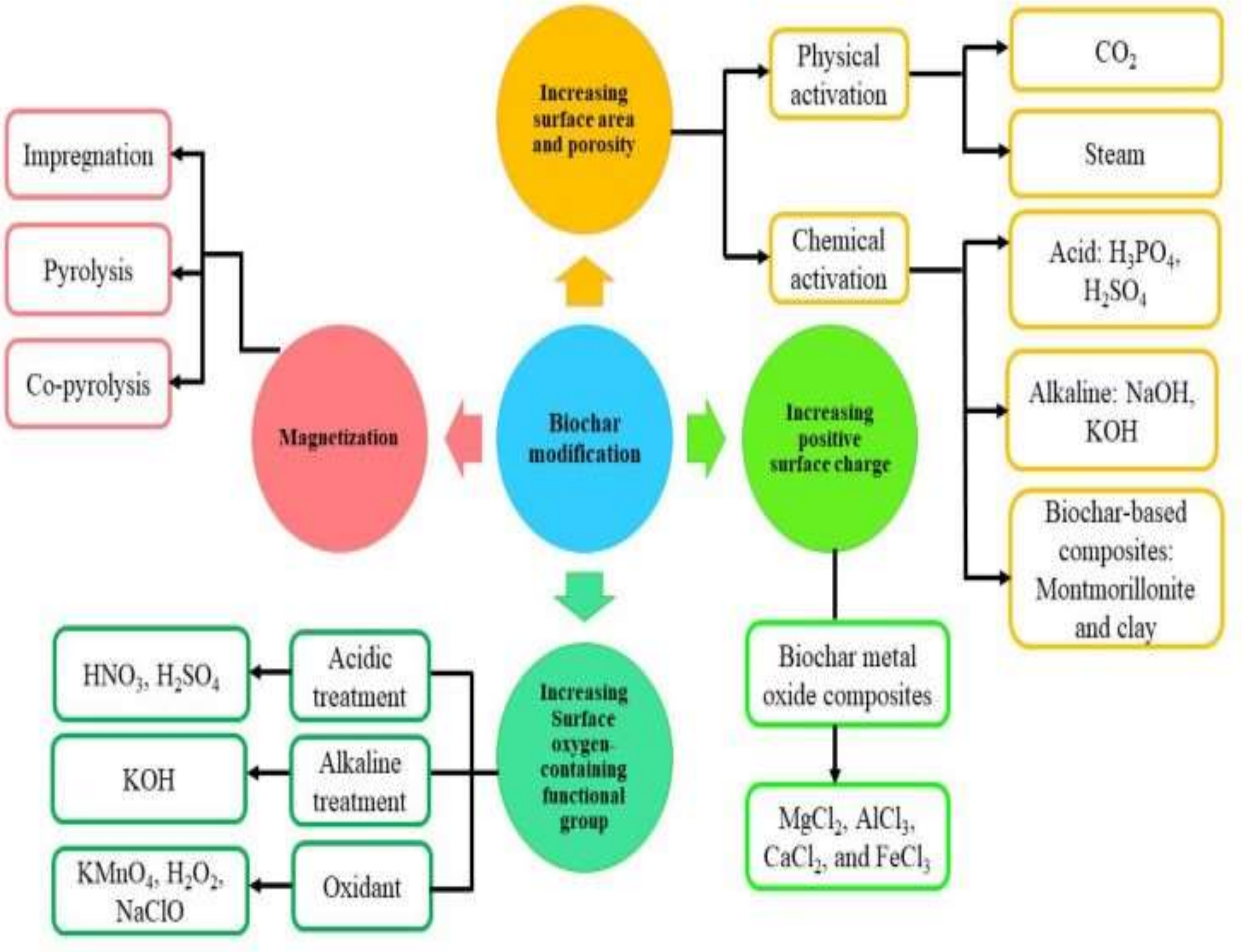

Fig. 3. Various modifications into the biochar (Adapted with permission from Ref. (Goswami et al., 2022)

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Table 2: Removal of antibiotics residues via adsorption by using different biochar

Material	**Antibiotics**	**Pyrolysis condition/ activating agents**	**Kinetics**	**Mechanisms**	**Ref**
Red pine	sulfamethoxazole	400°c	-	π-π electron-donor-accepter interaction	57
Alfalfa Medicago sativa L.	Tetracycline	500°c	PSOM	Surface complexation Hydrogen bonding Electrostatic interaction	58
Sugarcane base	Chlortetracycline	800°c	-	-	59
Rice husk	Tetracycline	H_2SO_4, KOH, 500-550°C	PSOM	Hydrogen bonding and π-π interaction	60
Cornhusk	Tetracycline Levofloxacin	$FeCl_3.H_2O$, 300°c	-	Hydrogen bonding and electrostatic interaction	61
Self-functionalised corncob biochar	Amoxicillin Tetracycline levofloxacin	Ultrasonic, 700°c	Elovich	Π-π interaction	62
Pomegranate wood	Amoxicillin	-	PSOM	Electrostatic interaction	63
KOH modified pomegranate peel	Ciprofloxacin	-	-	Hydrogen bonding and π-π interaction	64
Activated carbon from i. Banana peel ii. Straw iii. Avocado peel	Ciprofloxacin	-	-	electron-donor-accepter interaction and Hydrogen bonding and π-π interaction	65
Pumpkin seed derived activated carbon	ciprofloxacin	-	PSOM	Electrostatic attraction	66
ZnO-modified pistachio shells	Amoxicillin	-	PSOM	Chemisorption	67
Spent mushroom substrates	Sulfamethoxazole	-	PSOM		68

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Moringa oleifera	Oxytetracycline	-	PSOM	Electrostatic-attraction intra-particle diffusion	69
Banana peel graphene	Erythromycin		PSOM	N/A	70
Rice husk	Erythromycin		PSOM	N/A	71
Biochar from peanut shell	Chloramphenicol	105˚c for 12h	PSOM	Hydrogen bonding and π-π interaction	72
Garlic peel	Quinolone		PFOM, PSOM	Hydrogen bonding	73
Grape stalk	ofloxacin		PSOM	π-π interaction	74
Vine wood	Amoxicillin Cephalexin Tetracycline Penicillin G		PFOM and intraparticle diffusion	Protonation, Hydrogen bonding and vander walls forces	75

due to the aromaticity of the biochar, and polarity, with or without the needed/unneeded surface functional groups. Poor absorption, related to filling the pores, is caused by low porosity factors such as SBET, pore volume1, and limited diameter of the pores compared to the size of the antibiotic. It is essential to modify and improve the structure of biochars for increasing the adsorption capacity of modified BCs such as antibiotics. So it can be classified into the physical and chemical approaches (Krasucka et al., 2021). Fig. 3 shows different modifications of modified biochars for removing pollutants from the environment.

III. ACTIVATION METHODS OF BIOCHAR

i. Physical Activation

This approach includes activation by gas, ball milling, and microwave pyrolysis. The first two methods belong to the post-synthesis methods, in which modification is done on previously synthesized biochars. gas or steam activation and also ball milling is related to the post synthesis approaches. The third modification is using microwaves for heating during the pyrolysis and making modified biochars (Wang et al., 2017; Foong et al., 2020). After the treatment of bare biochar with above mentioned activating agents are referred as activated carbons(AC). Activaed carbon is a carbon product, which is obtained from agriculture waste; carbonaceous source material example like wood, peel, husk. It purportedly poses; large pore volumes, high adsorption capacity, adsorption favourable surface area, small pore diameter and have been worked as most efficient adsorbent. The properties of prepared AC depend on the preparation method and raw precursor materials. Various modification processes are depicted in fig. 2. They are more efficient, cost effective and having comparable better adsorption characteristics than commercially available Acs. Furthermore, to enhance the efficiency for antibiotics removal, modification in bare biochar by using graphene, carbon nanotubes, clay material, graphene oxides, metal oxides, hydroxides, and polymers has been employed to form biochar composites. These modifications can be done before pyrolysis or after pyrolysis according to requirement. However, the selectivity of adsorbents for target antibiotics plays an important role because it is quit bit challenging to know the type of contaminants present in the matrix.

ii. Chemical Activation

Post-synthesis chemical modifications such as oxidizing or reducing properties can be done on the pure biochar, and then it could be dried in simple ways (Wei et al., 2018) or advanced drying in the microwave (Ge et al.) 2020). In the pre-synthesis, raw material is chemically modified and then pyrolyzed (Wei et al., 2018). The difference between physical and chemical modifications is the major aim of physical modification is improving the porosity of biochar, while the main purpose of chemical modification is changing the essence of biochar, mostly causing the development in the surface oxygen functional groups (Sizmur et al., 2017). Table 2 shows the adsorption of antibiotics by using modified biochars.

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IV. ENVIRONMENTAL AND ECONOMIC ADVANTAGES OF BIOCHARS

The consumption of biochar as a pollutant adsorbent has brought a positive economic status. BC is a cheaper alternative to AC ($350-1200 per ton of BC versus $1100-1700 per ton of AC) (Thompson et al., 2016). The price of BCs relies on the cost of raw materials, pyrolysis, transport, and keeping the BCs (Shackley et al., 2011). The suitability of BCs in the economical aspect is very vital. Some data reports that the whole income from biochar sales was $8012 per year. By selecting new, cheap materials and optimal production technologies, the price of BCs could be decreased (Oni et al., 2019). Therefore, it is beneficial for the economy and the environment to use various kinds of waste and manure in the preparation of biochar (Oni et al., 2019; Ahmed et al., 2016). Finally, there are no doubt those BCs, as adsorbents for removing pollutants in water and wastewater, have positive impacts on the environment. It is necessary to ensure the harmlessness and reusability of the adsorbent. For this reason, the use of carbons for water treatment, relying on the application, must have the necessary standards, like European EN 12915 1:2009 (products used for water treatment intended for human consumption). According to the novel process of using BCs for waste water treatment, so far, no law has been established for their commercial use. Increasing the knowledge in the field of BCs increases efficiency and economic importance (Krasucka et al., 2021)

V. CONCLUSION

Environmental pollution with antibiotics has caused harmful and dangerous effects on humans, organisms, and the environment. The annual consumption of antibiotics is increasing sharply. This causes an increase in environmental pollution. As a result, studying and designing the most efficient approaches to eliminate this emerging pollution has garnered great attention. The outcomes illustrated that engineered biochar, as adsorbents, could be efficient for the removal of antibiotics. Biochar-based adsorbents can be effective for the removal of antibiotics by degradation. The absorption yield depends upon the type of composition and the absorption conditions and the characteristics of the adsorbent and the contaminant. All these factors affect possible absorption processes. The connection between the antibiotic with the BC adsorbent surface depends on the functional groups, the degree of BC graphitization, and pH. recognizing the connecting mechanisms for different antibiotics plays an important role in future research. With sufficient knowledge and information, it will be convenient to select an efficient and appropriate adsorbent to adsorb the desired antibiotic and to produce or optimize the required materials with a high yield of removal. Finally, the efficiency of biochar in removing emerging pollutants on a real scale is also an important requirement.

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