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TO STUDY THE REMOVAL OF PHARMACEUTICALS FROM AQUEOUS MEDIA BY ACTIVATED CARBONS

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ABSTRACT

Antibiotics are a particularly troublesome pharmaceutical pollutant, thus research into ways for removing them from water has attracted a lot of interest. Carbon-based compounds, such as activated carbon, carbon nanotubes, and graphene, are commonly considered to be among the most effective adsorbents for wastewater treatment and pollution management. Since they have a high surface area and interesting chemical and physical properties, adsorbents like activated carbon and carbon nanotubes are gaining in popularity for application in wastewater treatment. Adsorbents like activated carbon, carbon nanotubes, and graphene have showed promise in the study of organic and inorganic waste removal. Extraction of pharmaceutical active components from aquatic environments; production; benefits; uses of activated carbon; carbon nanotubes; and graphene.

Keywords:adsorption study, entrapped AG–AC, pharmaceutical wastewater, removal of COD

INTRODUCTION

When pharmaceutical waste such as ascorbic acid and lactose is disposed of in rivers and fields, it can promote the growth of viruses, bacteria, and other microorganisms that pose a serious threat to the environment or increase loading at the Processing Station, so these materials must be removed before they can be flushed down the drain. 4 Complex, costly, and time-consuming methods are used in wastewater treatment, including coagulation, flocculation, sedimentation, and aerobic and anaerobic biological contactor. For this reason, scientists have been working on a novel method for effectively removing organic matter from pharmaceutical effluent. 5–7 Although activated carbon has a powerful ability for adsorption8–12, its powdery form is notoriously difficult to eradicate from treated medium. Since activated carbon entrapped in alginate polymer facilitates and aids adsorption, and since it permits polluted aqueous solutions to come into contact with the entrapped activated carbon, this problem may be overcome. 10,13–19 This method of treating wastewater has gained a lot of attention in recent years since it is one of the most cost-effective methods available.

In addition to their widespread use in human medicine, antibiotics find widespread application in the animal husbandry and aquaculture industries as well. Recent studies show that the number of antibiotics used annually has grown dramatically, with the worldwide antibiotic consumption rate increasing by 39% between 2000 and 2015 owing to a rise in the recommended daily dosage of 65%. Although most antibiotics are effective, they are not digested in their entirety, resulting in the release of between 30 and 90% of the parent chemicals into the environment. Surface water, ground water, sea water, and soil have all been shown to contain antibiotics. Many surface waters have been shown to contain antibiotic residues, the most common of which are sulfonamides, macrolides, tetracycline (TC), and quinolones.

Due to their limited biodegradability, high persistence, and easy bioaccumulation, pharmaceuticals are now widely recognized as a significant developing contaminant. Humans and animals alike rely heavily on pharmaceuticals, sometimes known as medications, to enhance their health. Antibiotics, analgesics, anti-inflammatory medications, lipid regulators, hormones, and beta-blockers are all examples of widely used pharmaceuticals. Effluents from healthcare facilities, pharmaceutical manufacturers, and garbage dumps all include these biologically active compounds. Since many effluents from urban wastewater treatment facilities are polluted with PPCPs residues, their presence in the environment has attracted considerable attention in recent years owing to their pervasive usage in every facet of modern life. Most people agree that PPCPs pose a long-term threat to both ecosystem health and human health, and that their presence may reduce the quality of drinking water. In instance, tetracycline (TC), which is a kind of PPCPs, is widely used as both human and veterinary medication to cure ailments and promote growth. But it's not biodegradable and it's hazardous, and researchers have found TC concentrations as high as 86-199 g/kg in surface soil and 0.07-1.34 g/L in surface water. Clearly, it is crucial to find effective means of eliminating TC.

LITERATURE REVIEW

Chian Ying Teo et.al (2022) Antibiotics are a new kind of water contaminant that has garnered a lot of interest from scientists. Most antibiotics enter the environment via wastewater discharged from hospitals and factories, as well as through animal dung and agricultural fields that are irrigated. All living things are vulnerable to the dangers posed by antibiotic residues, the worst of which is the development of "superbugs," bacteria that are resistant to treatment with conventional antibiotics. When illnesses become resistant to antibiotics, treatment becomes more difficult, which is why this issue is of concern to the medical community. Therefore, it is important to create efficient and cost-effective methods for detoxifying the environment of antibiotics. Carbon-based adsorbents are basic materials that are well suitable for antibiotic adsorption, making adsorption a promising technique due to their efficacy and high operational practicality. In this article, we will summaries the present situation of antibiotic pollution, including the negative effects of various antibiotics and the difficulties inherent in antibiotic removal.

Aseel M. Aljeboree et.al (2018) Many scientists have studied methods for removing antibiotics from water because of their significance as a pharmaceutical contaminant. Considering that many pharmaceuticals are not quickly biodegradable and hence may survive and remain harmful in the environment, the detection of these substances in natural and wastewater bodies as well as drinking water has garnered a great deal of interest. Therefore, there is a continuing and prospective threat to human health and the environment from pharmaceutical residues. Antibiotics have found extensive usage with activated carbon (AC) as an adsorbent because to its large specific surface area, high porosity, and advantageous pore size distribution. The effectiveness of AC as an adsorbent for commonly used antibiotics as tetracyclines, Data showed that tetracyclines had the highest adsorption capabilities at 477.1 mg/g, indicating that AC adsorption was the most effective technique for treating these contaminants.

Mansouri F et.al (2021) Now more than ever, pharmaceuticals are essential medical supplies. Multiple studies have shown that drinking water contaminated with medications may have harmful consequences on people. Most medications in surface waterways and drinking water come from sewage and wastewater effluents. Thus, the efficacy of pharmaceutical removal during wastewater and drinking-water treatment procedures has to be considered and characterized. Drugs (e.g., antibiotics, NSAIDs, analgesics) may be removed or reduced using conventional or biological treatments such activated sludge processes or bio-filtration. Between 20% and 90%, these procedures may be considered efficient. High drug removal rates are possible with modern wastewater treatment methods including reverse osmosis, ozonation, and sophisticated oxidation technologies. Adsorption and photooxidation play a role in the gradual weakening of pharmaceuticals and their metabolites in the environment.

Chauhan, S., et.al (2022) One notable group of newly discovered pollutants is the pharmaceutical industry. There are several ways in which these biologically active molecules might harm people and ecosystems. This is because they are resistant to change, don't decompose very well, and are essentially permanent despite appearances to the contrary. The increasing use of pharmaceuticals in settings including hospitals, community health centers, and veterinary institutions has led to a dramatic rise in the prevalence of pharmaceutical residues in the environment. Several methods are being considered for their potential to rid water supplies of pharmaceutical chemicals (PCs). When compared to the others, adsorption stands out as the most practical treatment option due to its cheap cost and ease of usage. This has led to a surge in research and development towards the discovery of low-cost adsorbents for the efficient removal of PCs. Biochar-based adsorbents have attracted a lot of scientific interest to remove PCs from aqueous matrices because to their good specific surface area, variable surface chemistry, scalable manufacture, and ecologically benign nature, although many other adsorbents have been studied for this purpose.

A.Gil, et.al (2019) Six developing contaminants were studied in batch sorption studies using a commercial granular activated carbon as the adsorbent for aqueous solutions. We chose caffeine, clofibric acid, diclofenac, gallic acid, ibuprofen, and salicylic acid as our representative pollutants. Nitrogen adsorption at 77 K and the zero-charge-point measurement were used to characterize the activated carbon. Several operational factors were studied to see how they affected the sorption behavior; they included pH, starting organic molecule concentration, adsorbent mass, and contact time. Maximum adsorption equilibrium was reached after 40 minutes of contact time. Multiple adsorption models were used to explain the kinetic data, and it was determined that the adsorption process followed pseudo-second-order and intraparticle-diffusion models, with external mass transfer dominating in the first 15 minutes of the experiment. Several models, including the Freundlich, Langmuir, and Toth isotherm equations, were used to the equilibrium adsorption data.

TREATMENT TECHNOLOGIES FOR PHARMACEUTICAL COMPOUNDS' REMOVAL FROM WATER

Because of their existence in the aquatic environment at trace concentrations the efficacy of wastewater treatment systems in eliminating the pharmaceutical active chemicals has been called into doubt. Removal methods for pharmaceutical active chemicals from water bodies include chlorination, photocatalysis, adsorption, biodegradation, and advanced oxidation or ozonation. The high cost, high energy need, and the generation of harmful by-products are just a few of the many drawbacks of some of these methods. In comparison to these other methods, the adsorption approach has numerous benefits, including its ability to function in gentle operating circumstances, its low energy requirements, and its efficiency and cheap cost. As a result, this method has great potential for the elimination of pharmaceutically active substances.

Adsorption technique

Since adsorption is simple in design and operation, produces no harmful residues, and can effectively remove most kinds of organic material, it is widely regarded as one of the safest methods for extracting pharmacological active chemicals from water. The pharmaceutic substances accumulate on the adsorbent's surface as part of the adsorption process. Therefore, the adsorbent choice should be carefully considered. The adsorbent's large surface area and high hydrophobicity are necessary for removing the contaminant from the water. It is the functional group composition, surface area, pore size, and ash content of the material that determine how effective this method will be. The chemical factors, such as temperature, polarity, pH, adsorbate concentration, and the presence of other competing solutes, also have a role. Besides the adsorbent's active surface sites and the size of its pores, the adsorption process also rely on the ease with which the adsorbate molecules may move toward the adsorbent's boundary layer.

Activated carbon

Purified graphite in an amorphous, extremely porous state is what activated carbon is. It has holes ranging from microns in size to molecular-sized slits. In the early nineteenth century, wood was used as a raw material to make the first commercially produced activated carbon. Since 1930, it has been used to improve the flavor and aroma of drinking water. Sugar, shells, refinery coke, rice hulls, and various kinds of wood are only some of the primary organic materials and sources now used to create activated carbon. Activated carbon's primary advantages as an adsorbent are its... This is due to I its large surface area, (ii) its porosity, and (iii) its surface reactivity.

Classifications of activated carbons

Both the method of activation and the characteristics of activated carbon are used to categories the material. There are two primary groups delineated by activation procedure:

- Carbon that has been physically or thermally activated has undergone a carbonization process in which organic source materials are heated to temperatures between 500 and 600 degrees Celsius.
- Activating the surface of carbon requires the addition of inorganic ions like metallic chloride in the case of chemically activated carbon.

Physicochemical properties of activated carbon

The raw ingredients and activation technique used to create activated carbon have a significant impact on the final product's characteristics. Activated carbon may be altered in a number of ways due to its porous graphite and graphene sheets, which are linked together and contain -orbitals in the benzene rings. By way of illustration, the positive zeta potential of basic activated carbon may be changed to negative for usage in a variety of contexts by chilling the carbon in the presence of oxygen, which results in activated carbon rich with oxides and acidic functional groups. Furthermore, activated carbon's surface chemistry, pore structure (volume and diameter), and surface area are very temperature-dependent.

Activated carbon production

It is mentioned that a broad variety of raw materials may be utilized as a beginning material in the production of activated carbon. For the purpose of creating activated carbon, the following activation techniques are used:

- Thermal activation: this physical process might comprise two primary steps: first, carbonizing the raw materials at a temperature ranging from 500°C to 600°C to remove volatile substances; and second, improving the porosity and surface area by gasifying the material. High temperatures (between 800 and 1000 degrees Celsius) and an oxidizing gas (usually carbon dioxide (CO2), methane (CH4), or steam) are employed in the gasification process.

- To increase the micro-porosity and surface area of activated carbon, inorganic salts like metallic chloride are added prior to carbonization in the chemical activation process.

Activated carbon for removal of pharmaceutical active compounds

In adsorption operations, activated carbon (AC) is often employed as a filter and purification medium. To eliminate unpleasant odours and tastes, and to remove harmful metals and contaminants from water, activated carbon is employed in a variety of water treatment processes. Researchers looked at using AC to remove various pharmacological active chemicals because of its large surface area and commercial availability. Many of these drugs are summarized in Table 1. The antibiotic tetracycline, for instance, was removed from aqueous medium using several forms of activated carbon. Using macadamia shells as a source, Martins et al. generated activated carbon with a yield of 19.79% and a surface area of 1524 m2/g. It was utilized to get rid of tetracycline and had an adsorption capability of 455.33 mg/g. As reported by Muthanna et al., activated carbon was used to remove tetracycline, penicillins, and quinolones from a solution, and the activated carbon they employed had an adsorption capacity for tetracycline of 1340.8 mg/g.

Carbon nanotube

Materials known as single- and multi-wall carbon nanotubes (CNTs) are essentially graphene sheets that have been coiled up into tubes. Using the arc discharge technique of manufacturing and subsequent characterization using a transmission electron microscope, CNTs were first found by SumioIjima in 1991 at NEC Laboratory in Japan. Figure 1 shows that CNTs may take on two distinct shapes depending on the rolling orientation of their graphene sheets.

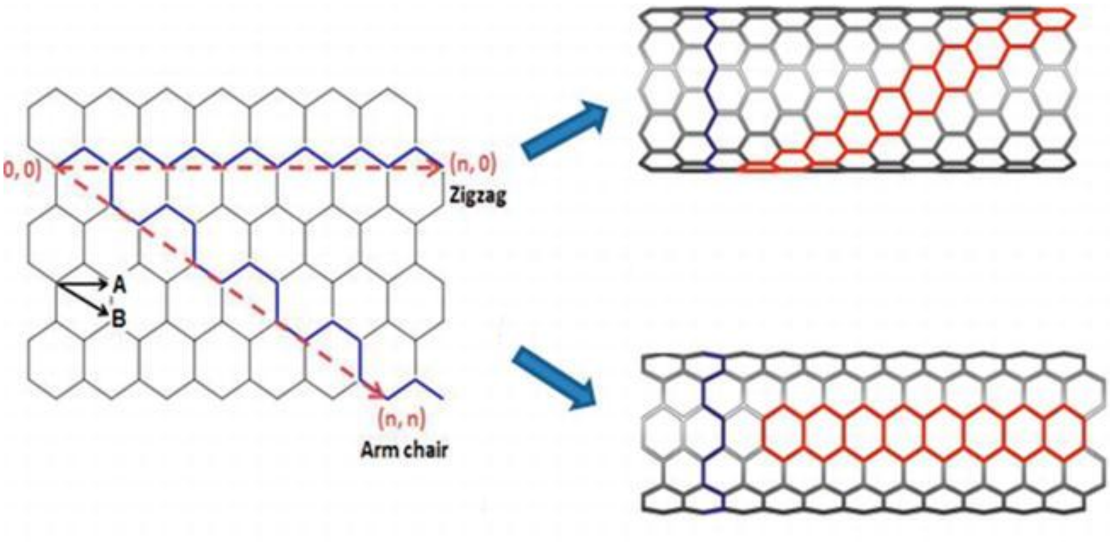

Figure 1.Carbon nanotube

Armchair and zigzag structural forms of CNTs

Cylindrical CNT nanostructures may have a length to diameter ratio of up to 132,000,000:1, making them the longest and thinnest materials yet created. The sp2 hybridization of the carbon atoms that make up CNTs, together with their inherent alignment into ropes drawn together by Van der Waals attraction, accounted for this characteristic.

Physical properties and chemical reactivity of carbon nanotubes

CNTs group together to create a very intricate web. The hexagonal ring configuration along the tube's surface determines the material's electrical conductivity. CNTs are appealing candidates in many nanotechnological applications, including the elimination of pharmaceutical chemicals in water treatment procedures, because of their exceptional features, such as huge geometric aspect ratio, nanocavities, and electrical conductivity. A lack of excellent suspension characteristics in aqueous and organic solvents is a major limitation of carbon nanotubes (CNTs), which has hampered their usage in industry. Chemically altering CNTs with certain hydrophilic functional groups improves their suspension in water, mitigating this drawback.

The main distinct properties of the carbon nanotubes are categorized into the following:

Mechanical properties: CNTs are very strong and rigid because of the covalent sp2 connections created between individual carbon atoms. Results show that carbon nanotubes (CNTs) have an elastic modulus of 1Tpa, which is orders of magnitude more than that of steel (10-100 times greater). Table 1 displays a comparison between CNTs and a variety of materials known for their high-quality mechanical characteristics.

Table 1.the carbon nanotubes are categorized

Physical property	Material name				
	MWCNTs	SWCNTs	Wood	Steel	Epoxy
Density (g/cm^3)	2.6	2.6	0.6	7.8	1.25
Tensile strength (Gpa)	150	150	0.008	0.4	0.005
Young's modulus (Gpa)	1200	1054	0.6	208	3.5

Examination of CNTs in relation to other materials.

- The thermal conductivity of CNTs is between 2800 and 6000 W/m K.
- Carbon nanotube (CNT) based materials may be either a conductor or a semiconductor, showcasing their exceptional electrical characteristics. Thousands of times more electricity can be carried by the conductive CNTs than by copper.
- CNTs may be chemically changed to increase their efficiency and make them more soluble in aqueous and organic liquids. The curvatures in CNTs' structure led to a mismatching of -orbitals, which contributes to their reactivity. Overall, the reactivity of nanotubes increases as their diameter decreases. Chemical alteration of CNTs' sidewalls or end caps was also shown in the presented findings.

The aforementioned features of CNTs suggest that CNT materials and their modified structures have great promise in fields as diverse as water treatment, environmental protection, pharmaceutical active component removal, material science, medicinal chemistry, and others.

Carbon nanotube production

Several methods exist for creating CNTs, the most popular of which are:

- The use of an arc discharge method. The simplest and most often used method for creating CNTs is the arc discharge method. The first CNTs were found using this method. By using an arc discharge with an inert gas like helium or nitrogen, CNTs may be manufactured at low pressure. They are made by arc vaporising two carbon rods that are kept apart in an inert gas environment. The created CNTs are not pure since they retain part of the catalytic metals; hence, they must be purified to eliminate these metals and acquire clean CNTs.

• Ablation using a laser. Carbon nanotubes were first manufactured in 1995 by vaporizing graphite at 1200 degrees Celsius using a laser beam. Pulsed laser techniques and continuous laser ablations are the two most common. The primary distinction between the continuous and pulsed laser ablation techniques is the much greater light intensity (100 kW/cm2) utilized in the former. The laser ablation technique involves vaporizing carbon and then collecting the resulting CNTs on a colder surface inside the reactor. By including metal-based catalysts like Co, Fe, and Ni into the solution, SWCNTs may be synthesized from graphite electrodes in this method. When a pure graphite electrode is used, however, MWCNTs are the primary byproduct.

• Chemical vapor deposition (CVD) method. The chemical vapor deposition method is seen to be the simplest approach to mass-producing CNTs in industry due to its straightforward nature. By adjusting the system's production parameters including temperature, catalyst, and carbon supply gases, the required CNT type and quality may be manufactured. There are two primary phases to the CVD procedure (catalyst preparation step and then CNT synthesis). When making CNTs, it is common practice to split methane and carbon monoxide gases into reactive carbon atoms using an energy source, and then to have these reactive atoms diffuse across a substrate coated with transition metals as a catalyst and heated to between 500 and 1000°C.A comparison between the previously discussed methods for CNT production is summarized in Table 2.

Table 2.Carbon nanotube production

Method	Yield (%)	SENT	MINT	Concerns
Chemical vapor deposition	20–100%	Long tubes with diameters 0.6–4 nm	Long tubes with diameters 10–240 nm	Nets are usually mints and often riddled with defects
Arc discharge	30–90%	Short tubes with diameters 0.6–1.4 nm	Short tubes with inner diameter 1–3 nm	Short tubes with random sizes and directions and required purification
Laser ablation (Vaporization)	Up to 70%	Long bundles of tubes with diameters 1–2 nm	Not suitable and too expensive	Costly and required high power

In this comparison, we will look at the present drawbacks of each technique, as well as the efficiency with which it produces CNTs and the many types of CNTs it can generate.

Carbon nanotubes for pharmaceutical active compounds' removal

Carbon nanotubes, because to their exceptional characteristics, have significant adsorption potential for removal of pharmacological active substances. Single wall carbon nanotubes (SWNTs) were shown to be more effective in removing tetracycline from aqueous solutions than multiwall carbon nanotubes (MWNTs), graphite, and activated carbon, according to research published in 2009. It was determined that the molecular sieving effect was responsible for this observation, since the large molecules of tetracycline were unable to permeate the interior pores of the adsorbent. Adsorption performance of MWNTs for removal of ciprofloxacin was investigated by Yu et al. in 2016, and it was found that the maximum adsorption capacity is 20 mg/g, obtained at pH 4 and 240 min, and attributed to the - interaction rather than hydrogen bonding and interaction with oxygenated functional groups on MWNTs. Maximum tetracycline adsorption capacity of MWNTs was 269.54 mg/g, as shown in another work by Yu et al., which took just 80 minutes to reach at 25 degrees Celsius and pH 5.

Numerous changes, including graphitization, hydrolyzation, carboxylation, and etching with potassium hydroxide, may be applied to CNTs to enhance their performance and adsorption capacity Ji et al., for instance, etched SWNTs and MWNTs with KOH and then evaluated the resulting CNTs for three pharmacological active chemicals KOH modified single-walled carbon nanotubes and multi-walled carbon nanotubes showed improved adsorption performance for sulfamethoxazole (56%) and tetracycline (84%), respectively, compared to unetched SWNTs and MWNTs. The increased surface area of the etched CNTs has been proposed as a possible explanation.

Graphene

Graphene is a nanomaterial consisting of a single sheet of sp2 hybridized carbon atoms, and it is two dimensions in size. Graphene is a nanomaterial with remarkable qualities, such as a high specific surface, high electrocatalytic activity, strong thermal conductivity, high stiffness and strength, and a high electron mobility. Scientists were first intrigued by its peculiar physical features, which led to its introduction for a variety of possible uses. Adsorptive removal of developing contaminants, such as pharmaceutical active chemicals, is one example.

Graphene for pharmaceutical active compounds' removal

Because of their high surface area and catalytic activity, graphene nanomaterials and their modified forms have a wide range of potential uses, including the adsorption and removal of pharmacological active chemicals. The effectiveness of graphene oxide as an adsorbent for the removal of the antibiotic tetracycline from water was studied. Researchers determined that the maximal monolayer adsorption capacity for tetracycline is 313 mg/g, and that this value decreases with increasing solution pH or sodium ions concentration. These substances included carbamazepine, sulfamethoxazole, sulfadiazine, ibuprofen, paracetamol, and phenacetin. Adsorption capabilities were determined to be 340.5 mol/g, 428.3 mol/g, 214.7 mol/g, 224.3 mol/g, 350.6 mol/g, and 316.1 mol/g for these chemicals, respectively. Adsorption of acetaminophen, aspirin, and caffeine from aqueous solution using graphene nanoplates (GNPs) was shown to be spontaneous and exothermic in 2014 research, with corresponding adsorption capacities of 18.07 mg/g, 12.98 mg/g, and 19.72 mg/g.

One of the primary drawbacks of employing graphene as adsorbents is that its surface area decreases dramatically in solutions owing to its aggregation, lowering its adsorption capability. Adding a functional group or metal to graphene is one way to improve its adsorption capability and counteract this drawback. We have explored the adsorptive removal of four tetracycline (TC) pharmaceutical active chemicals (i.e. tetracycline, oxytetracycline, chlortetracycline, and doxycycline) from aqueous solution by functionalizing a graphene oxide with magnetic nanoparticles. Maximum adsorption capacity for TC was determined to be 39.1 mg/g, with the solution pH and ionic strength having little to no influence.

The adsorption removal of antibiotics by activated carbon Adsorption removal of tetracycline

Because of its enormous surface area and strong adsorption ability for a wide variety of pollutants, AC has been employed as an adsorbent. The evolution of activated carbon may be broken down into two main categories: the quest for low-cost raw materials and efficient activation techniques, and the enhancement of adsorption capacity via doping active carbon with other inorganic chemicals to activate them in diverse ways.

SEM images of ITAC (Iris tectorum with H3PO4) and ITAC-Fe (Iris tectorum with ferric nitrate) were discovered, as well as images of the manufactured ferric nitrate-doped activated carbons (the mixture of H3PO4 and ferric nitrate). Adsorption of TC was shown to be highly pH-dependent, with adsorption ability dropping off precipitously when solution pH was raised. TC may absorb up to 769 milligrams per gramme.

resulted in A high yield (19.79%) and surface area (1524 m2 g1) for tetracycline were observed in activated carbon (AC) made from macadamia nut shells, as was a sizable adsorption capacity (455.33 mg g1).

Using a modified graphite felt as the cathode in a flow-through electro-Fenton (EF) and in-situ regenerative active carbon felt (ACF) adsorption system, they showed Tetracycline (TC) could be removed economically. It shown that the coupling process maintained steady performance during the whole 600 min treatment, resulting in a removal efficiency that was 5-44% greater than that of a single flow-through EF system.

Tetracycline (TC) was successfully removed from water using activated carbon (AC) made from apricot pits and phosphoric acid. Total pore volume was 0.191 cm3, average pore width was 1.957 nm, and specific surface area was 1.957 nm,307.6 m2 g1. Adsorption of temperature and concentration was investigated as a function of contact duration, adsorbent dose, starting TC concentration, and initial pH of the solution.

CONCLUSIONS

Removal of pharmaceutical active compounds from aquatic media can be achieved by either conventional or advanced methods. Among them, the adsorption technique has many advantages over the others. Several materials as adsorbents have been reported and discussed in the literature such as silica-based adsorbents, polymeric materials, clay, carbonaceous materials and other materials. Activated carbon, carbon nanotube and graphene oxide among carbonaceous materials show excellent performance and high adsorption capacity for pharmaceutical active compounds. As discussed in this chapter, the activated carbon can be activated using different methods (i.e., physical or chemical activation), while the carbon nanotube can be produced through using one of the following methods: (i) arc discharge, (ii) laser ablation and (iii) chemical vapor deposition. The physical (surface area and porosity) and chemical (functional groups) properties are significantly affected by the followed production method for these carbonaceous materials.

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