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Nature's revelation: Preparation of Graphene-based Biomimetic materials and its application prospects for water purification

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ABSTRACT

Pollutants contained in wastewater pose serious harm to the environment. Graphene-based water treatment materials show significant advantages in wastewater treatment. However, with the development of graphene-based materials, its progress in water treatment has reached a bottleneck. The challenge lies in effectively enhancing its performance in water treatment and ensuring its practicality. By employing biomimetic approaches, some exceptional properties and structures found in nature can be mimicked in graphene materials, effectively enhancing graphene's adsorption and mechanical properties. Current biomimetic methods include biomimetic mineralization, self-assembly, and templating. Unfortunately, all of the above methods suffer from the disadvantages of complexity and poor bionic effect. Nevertheless, 3D printing, a form of additive manufacturing (AM) technology, offers integrated molding and excellent biomimetic performance in creating biomimetic materials. This paper will cover the following aspects: (1) An overview of objects suitable for bionics in terms of functional and structural aspects, along with their properties, and a discussion of various bionic objects combined with graphene materials in water treatment and related research; (2) a comparison of different methods for preparing graphene-based bionic materials; (3) an examination of the current drawbacks and limitations of graphene-based biomimetic materials; and (4) a conclusion and future prospects, exploring the potential of using 3D printing technology to produce graphene biomimetic materials. This review aims to serve as a guide for effectively leveraging natural inspirations to create graphene-based biomimetic materials and enhance graphene properties.

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1. Introduction

The term biomimicry, first proposed by the scientist Otto Schmidt [1], is an effective way to simulate the function, form, and performance of natural substances [2], and biomimicry can be simply divided into (1) physical structure bionics [3,4] and (2) functional bionics [5]. Structure bionics is applied to new materials by imitating some specific forms or structures in nature, focusing on imitating the existing structures [6]. Functional bionics is through the imitation of some biological predation [7], respiration [8], and some other physiological characteristics so that some materials in the application, with the same or similar functions, which are characterized by certain methods and means, mainly to achieve some

specific functions of nature [9]. Whether it is structure or functional bionics, they are inspired by borrowing the existing nature, some of the nature of the original nature and function, integration, and application to the study of materials and equipment [10]. Bionic materials have been widely used in aerospace [11], medical [12], architecture [13], and production and life, providing an irreplaceable role for research and development in various fields. It is essential to study the relationship between the structure of natural organisms, the mechanism of action of organisms, and the performance of the organisms themselves by studying the structure of the organisms and applying this relationship to the materials under study.

Adsorbent materials have been widely used in water treatment due to their excellent properties, such as simplicity, efficiency, and economy, and have received extensive attention in water treatment [14,15]. Therefore, in recent years, researchers have developed dif-

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ferent types of adsorbent materials, such as carbon nanotubes [16], biochar [17], sludge materials [18], and 3D graphene [19]. However, the new adsorbent materials developed have overcome some of the drawbacks of previous adsorbent materials. Such as improved adsorption capacity can be separated from water more efficiently [20]. However, with the development of adsorbent materials, how to construct the internal microstructure of the materials more rationally and develop the adsorption performance of adsorbent materials with the maximum potential has become an urgent problem that needs to be solved. These problems have brought the development of adsorbent materials to a certain bottleneck and hindered the development of adsorbent materials. The inspiration to solve these problems can be well obtained in nature, and the bionic approach provides an excellent way to solve them [21]. There are two main aspects: (1) The distribution of suitable macropores, mesopores, and micropores, and the relationship between the microstructure and the shape of the adsorbent seriously affect the adsorption and mechanical properties of adsorbent [21,22]. Too many macropores increase the material's brittleness, while too few macropores affect the mass transfer of the adsorbent material. Micropores affect the adsorption performance of the adsorbent material [23], while the microstructure and the appearance of the structure affect the specific surface area of the material and the mechanical properties of the material. By studying the original porous structure in the natural environment, researchers have found that the naturally occurring porous structure possesses a reasonable distribution of macropores, mesopores, and micropores. These reasonable pore size distributions not only greatly enhance the mechanical properties of the material [24] but also make the mass transfer of pollutants inside the material more pronounced, enhancing the material's adsorption performance. (2) In addition, some specific functions of nature are also the key to bionics [25]. Different surface properties, such as the presence of different functional groups and the balance between functional anions and cations, affect the capture of heavy metal ions [26]. These bionic ideas can be due to getting inspiration to better apply to adsorbent materials and break through the bottleneck of adsorbent materials to provide a reference for researchers to study adsorbent materials. For example, by imitating the hydrophobicity of the surface of lotus leaves, researchers have prepared hydrophobic materials with different characteristics.

Graphene is a sp^2 hybridized carbon atom tightly packed to form a two-dimensional honeycomb lattice structure material [27]. Graphene has a large specific surface area, excellent mechanical, electrical, and acoustic properties, and fascinating properties for water treatment adsorption [28–30]. Researchers have developed graphene materials from two dimensional (2D) to three-dimensional (3D) [19], a measure that has dramatically improved the applicability of graphene materials in water treatment. However, with the development of 3D one-piece macroscopic body graphene, the adsorption performance reaches a bottleneck due to the limitation of mass transfer in the internal pore size of 3D graphene and the surface properties. While most of the current research on graphene focuses on 3D printing graphene bulks [31], more studies are needed on combining graphene and bionics techniques. The future will focus on how to mimic the existing structures and functions in nature and apply them flexibly to graphene materials.

Although bionics have been widely used in recent years and developed in different fields, they have yet to be applied to water treatment. The research on bionic water treatment adsorbent materials is relatively few at present, while the research on the use of some excellent properties already existing in nature, and thus the preparation of 3D graphene bionic adsorbent is even fewer, which is not able to effectively provide a reference for the application of bionic materials in the field of adsorption and to provide re-

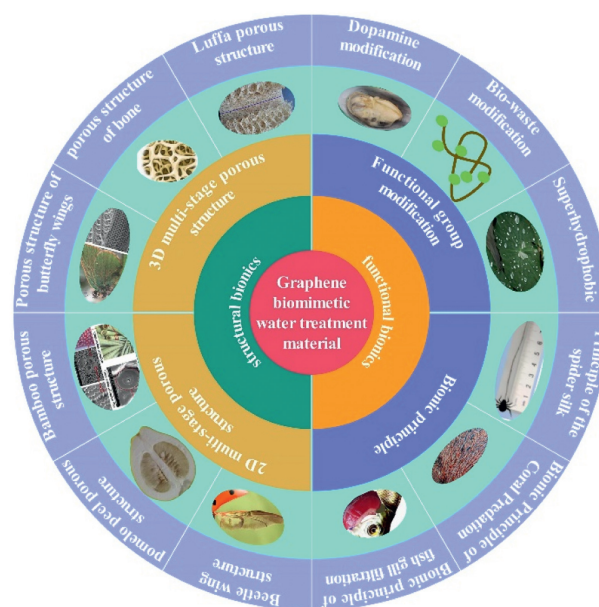


Fig. 1. Classification and description of bionic materials.

searchers with a practical idea to draw on. This paper aims to provide the research progress of biomimetic adsorbent materials for water treatment, including the existing biomimetic objects as well as the current research status, while describing the existing preparation methods of graphene biomimetic materials, as well as the current status of the biomimetic research, and finally reviewing the research and application of graphene-based biomimetic adsorbents for water treatment. In addition, the current bionic preparation methods include bionic mineralization, template, self-assembly, etc. However, most of them have the disadvantages of unsatisfactory bionic effect, cumbersome operation, and poor integrality. Compared with the other methods mentioned above, the 3D printing method has unique advantages in preparing graphene bionic materials. 3D printing technology has the advantages of integrated molding, precisely controllable printing, and good bionic effect. 3D printing has been widely used in the design of bionic materials, but there is currently little related research being applied to the water treatment field. Therefore, this paper summarises the research on graphene bionic adsorbent materials from two perspectives: functional and structural bionics. Different biomimetic preparation methods are described, and the research on the preparation of graphene biomimetic adsorbents by 3D printing technology is prospective. This paper describes graphene materials, biomimicry, and 3D printing technology in an integrated manner, which is expected to provide a reference for further developing graphene water treatment materials.

2. Bionic materials

Bionic materials have, in recent years, been widely developed and applied [32]. Bionic technology can effectively improve the performance of materials without additional treatment, which traditional methods cannot directly achieve. Similarly, replicating natural inspirations into graphene water treatment materials can improve their performance. Fig. 1 and Table S1 (Supporting information) show that the graphene bionic water treatment adsorbent materials are classified and summarised.

2.1. Functional bionic material

Some natural plants and animals show excellent properties and functions in some aspects, which are of great significance in bionics. Functional biomimetic materials are mainly based on mimicking some specific functions of nature; for example, researchers have prepared bioadhesives inspired by the high viscosity and strength of mussel secretion [33]. In addition, mussel biomimicry has been used in different applications in biomimetic research in medicine and the environment [34]. As another example, researchers found that the surface of the lotus leaf has excellent self-cleaning properties through research, and based on this excellent function, bionic materials with different effects of self-cleaning properties were prepared. The fascinating properties of functional biomimetic materials go far beyond that, and many inspirations in nature need to be constantly explored. The following section provides an overview of different functional bionics objects in nature and describes their potential for application in the field of graphene water treatment adsorption and the current state of research.

2.1.1. Lotus leaf effect

In 1977, Barthlott and Neinhuis of the University of Bern, Germany, investigated the surface structural morphology of the lotus leaf by scanning electron microscopy. They proposed that the micron-sized papillae and waxes on the surface of the lotus leaf are the key to its superhydrophobicity, which keeps the water droplets on the surface of the lotus leaf in the shape of a sphere and gives it excellent superhydrophobicity. This phenomenon is also called the lotus leaf effect or the lotus flower effect (Fig. S1 in Supporting information). By utilizing its properties, self-cleaning, antibacterial, and oil-absorbing materials can be effectively prepared. Nowadays, this superhydrophobic phenomenon is also applied to graphene materials. By mimicking its properties, superhydrophobic graphene-based materials are prepared.

For example, Li *et al.* [35] reported a superhydrophobic material based on a mixture of graphene oxide (GO) and carbon nanotubes (CNTs) to mimic a lotus leaf. Wang *et al.* [36] prepared high-output friction nanogenerators (TENGs) by doping graphene into structured polydimethylsiloxane by mimicking the self-cleaning property function of lotus leaves; similarly, Wang *et al.* [37] inspired by the hydrophobicity of *Oxalis corniculata* Linn in nature, prepared superhydrophobic laser-induced graphene (LIG) *in situ* for the first time by biomimetic means. In addition, Han *et al.* [38] developed a superhydrophobic chip with integrated lotus leaf effect and rose petal effect (LLE-RPE) on polyimide film using graphene. At present, the function of superhydrophobicity represented by the lotus leaf effect has been widely applied to graphene materials, mainly to take advantage of the self-cleaning property brought by the superhydrophobicity function.

However, due to the difference in the wettability of bionic superhydrophobic material from water and oil, it can be well used in water treatment adsorption. By taking advantage of graphene's developed pore structure and its selective adsorption of oil and organic solvents on the surface, it is possible to deal with oil contamination on the surface of water bodies and underwater. Therefore, researchers have done many studies on this subject.

For example, Xu *et al.* [39] synthesized an aerogel based on graphene oxide, nanofibres, and polyvinyl alcohol materials in a simple and environmentally friendly way to solve the oil pollution events. The aerogel had a water contact angle of 156°, which resulted in an excellent oil adsorption capacity of 97 times its weight. Mao *et al.* [40] decorated silica/graphene oxide wide ribbons (GOWR) onto a melamine sponge skeleton, followed by surface modification of the silica nanoparticles, and prepared composites with highly efficient oil adsorption efficiency. In addition,

the different densities of oil affect the adsorption performance of graphene-based biomimetic superhydrophobic materials. Therefore, Yang *et al.* [41] prepared super-hydrophilic/underwater super-oleophobic graphene oxide composites (GO@MS), as well as reduced graphene oxide/melamine sponge composites with super-hydrophobic/super-oleophilic (rGO@MS) by dip-coating GO on melamine sponges (MS) with chemical reduction treatment. GO@MS and rGO@MS have high selective absorption capacities for oil and water, with adsorption capacities in the range of 72.3–136.5 g/g. In addition, GO@MS and rGO@MS have good recovery properties and exhibit a high separation efficiency of 99% after compression. The biomimetic application of superhydrophobic functionality can be applied not only for the adsorption of oil contamination but also for pharmaceutical contamination. Such as, Akpotu *et al.* [42] first synthesized graphene oxide using the Tours method, followed by the introduction of methoxy ether polyglycerol (mPEG) onto the surface of graphene oxide. Finally, GO-mPEG was immobilized on a nanoporous material (SBA-15), and a superhydrophobic material (SBA-rGO-mPEG) was prepared by *in situ* homogeneous reduction, which efficiently adsorbs estrogen in water.

In general, the lotus leaf effect, a superhydrophobic phenomenon, has attracted extensive attention from researchers since its discovery. Scientists have prepared graphene materials with superhydrophobic functions through various means. By mimicking its function, the application of graphene materials in the field of water treatment adsorption has been extended.

2.1.2. Mussels and dopamine

Dopamine (DA) is a neurotransmitter containing catecholamines, and inspired by mussel adhesion proteins, researchers have found that dopamine can oxidize and self-polymerize to produce polydopamine (PDA) under alkaline conditions. Polydopamine has the following advantages: (1) Excellent adhesion, which can cover any surface and form a stable and uniform membrane layer; (2) wealthy functional groups, which have abundant catechol and amine functional groups that can become the active sites of heavy metal ions through electrostatic interactions, hydrogen-bonding interactions or bidentate chelation. These advantages make dopamine and its product polydopamine have unique advantages in preparing functional biomimetic materials for water treatment. For example, Gao *et al.* [43] prepared a novel biomimetic adsorbent material inspired by mussel secretion. A layer of PDA was first deposited on the surface of layered double hydroxide, followed by forming a metal polyphenol network of Fe^{3+} on the surface of PDA, resulting in a secondary structure. The presence of the secondary structure increases the adsorption sites and makes the adsorbent material highly selective for malachite green (MG) and crystal violet (CV).

In addition, dopamine substances have an excellent interaction with graphene materials. So that dopamine can be applied well to graphene by bionic means. Dopamine is a concatenate bridge to graphene, which plays a vital role in enhancing the stability of graphene sheets. Dopamine, inspired by mussel-secreted proteins, plays an interactive role with graphene through polymerization. This interaction not only enhances the stability of 3D graphene but also improves its adsorption properties.

Therefore, researchers have done much research on this to achieve the functional bionics between dopamine and graphene and its application in water treatment. For example, Song *et al.* [44] developed an ultralight, three-dimensional (3D), and nitrogen-doped graphene aerogel (NGA) using dopamine. DA undergoes self-polymerization, which can functionalize the graphene surface and embed nitrogen atoms into the graphene sheets upon pyrolysis. NGA not only possesses stable mechanical properties but also very high adsorption capacity and efficient space utilization of the sorbent space, which promotes the practical application as an oil ab-

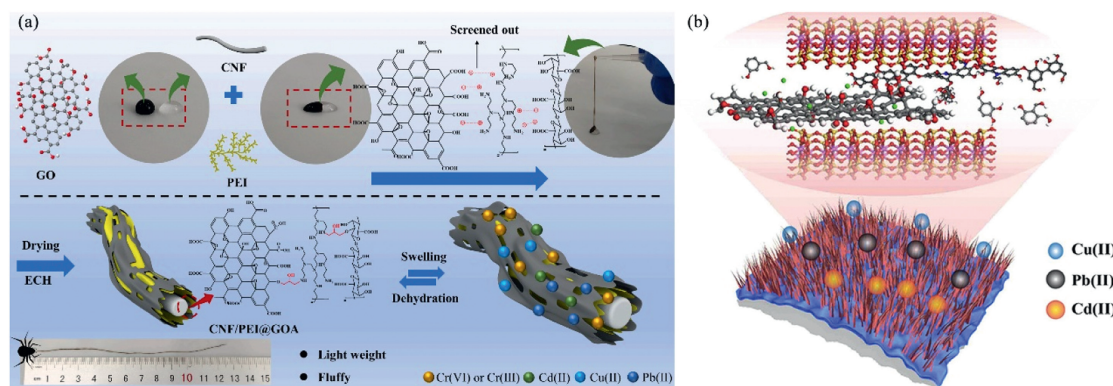


Fig. 2. (a) Adsorption mechanism of spider silk bionic material. Reproduced with permission [26]. Copyright 2021, Elsevier. (b) Adsorption mechanism of coral bionic material. Reproduced with permission [7]. Copyright 2020, Elsevier.

sorber for recovery of oil spills or chemical leaks at sea. Similarly, Zhang *et al.* [45] reported an aerogel (MWCNT-PDA-CS-GO) based on PDA, graphene oxide, chitosan (CS), and carbon nanotubes (MWCNT), which adsorbed up to 150.86 mg/g of Gd(III) under neutral conditions. To improve the adsorption performance of graphene materials for antibiotics, Yu *et al.* [46] used dopamine hydrochloride and β -cyclodextrin to modify graphene oxide. Moreover, its adsorption performance was evaluated by adsorption experiments on sulphonamide antibiotics; the results showed that the maximum adsorption amount reached 152 mg/g, which has good adsorption performance.

Due to the narrow layer spacing of graphene oxide, composite membranes of GO tend to suffer from the drawback of lower water flux in water treatment. To solve this problem, Ma *et al.* [47] prepared biomimetic dopamine/reduced graphene oxide composite membranes by modification of graphene oxide using PDA followed by vacuum filtration. The results showed that the stability of the composite membrane was enhanced due to the cross-linking effect of dopamine; in addition, the composite membrane had a better removal of dyes and heavy metal ions from wastewater due to the biomimetic modification of dopamine.

2.1.3. Spider silk

Spider silk is a natural macromolecular fiber with the advantages of high tensile strength, high tenacity, high thermal conductivity, and super shrinkage. In addition, spider silk is amphiphilic in aqueous solution, which makes it uniquely useful as an amphiphilic adsorbent. Researchers have made different explorations to copy the spider silk's function and its application in graphene adsorbent materials for water treatment.

For example, to solve the problem of the last kilometer removal of heavy metal ions from water, Zhou *et al.* [26] proposed that by mimicking the amphiphilicity as well as the stability function of spider silk, thus enabling the preparation of graphene adsorbent materials (CNF/PEI@GOA) with high adsorption performance and excellent stability. It was firstly assembled by electrostatic assembly with graphene oxide and polyethyleneimine (PEI) and then combined with a quartz crystal microbalance to precisely regulate the charges of carboxyl and amino groups, which led to the equilibrium of -COOH , NH_4^+ . The rapid and effective removal of heavy metals such as Cd(II), Cr(VI), Cu(II), and Pb(II) at a starting concentration of 1000 ppb was achieved. The high adsorption performance of the prepared bionic spider silk is fundamentally attributed to (1) the high density and equilibrium of amphiphilic functional groups and (2) the porous structure of the bionic spider silk with excellent mass transfer. Fig. 2a demonstrates the construction process of bionic spider silk and describes the intrinsic

role of electrostatic action, chelation, and adsorption of heavy metal ions.

Similarly, Wei *et al.* [48] utilized the special conformation of spider silk. Silica polystyrene fibers, encapsulated on polymethyl methacrylate fibers, were fused together using a high-voltage electric field. The prepared composites had a contact angle of 162° to water, which provided excellent filtration of water-in-oil solutions, currently, in addition to mimicking the properties of substances such as lotus leaves, spider silk, and mussels and applying them to water treatment materials such as graphene. Researchers have also studied the predatory role of corals (Fig. 2b) and applied it to graphene bionic water treatment materials. However, related research needs to be explored further.

In conclusion, functional bionic research has good potential and role in the field of water treatment, especially in the field of graphene water treatment adsorption. The current graphene functional bionic water treatment adsorbent material research can draw on the theory of the lotus leaf effect theory, spider spinning principle, and coral predatory behavior. These bionic theories are well-referenced in the field of graphene water treatment adsorption. The essence of the role is that the enrichment of the functional groups on the surface of graphene or making graphene adsorption has a selective, and thus enhances its adsorption performance. At present, there is more research on the preparation of graphene water treatment adsorbent materials by functional bionics, which is mainly due to the following two points: (1) The means and methods to achieve specific functions in nature are not limited; (2) The threshold of functional bionics is low. Meanwhile, functional bionics is important in improving the adsorption and related properties of graphene. Therefore, to effectively promote the development of graphene-based functional biomimetic water treatment materials, different kinds of natural substances can be explored and borrowed, and their properties can be imitated and applied to graphene as well as other water treatment materials. The water treatment performance of graphene can be greatly improved, and its application potential increased.

2.2. Structural biomimetic materials and multistage porous structures

Structural bionic materials are used for specific purposes by mimicking specific structures already existing in nature [3] to realize structures that cannot be directly realized by traditional methods [10,49,50]. Structural bionic materials have more complete and thorough characteristics than functional bionic materials. In recent years, the research of structural bionics has been widely applied in aerospace, new materials, medicine, and other fields [51,52]. For example, sharkskin's microscopic groove structure, which can effectively inhibit and delay the occurrence of turbulence, thus re-

ducing the resistance [53,54], can be effectively applied in drag reduction.

Some unique structures can also be borrowed in the study of adsorbent materials for water treatment, such as hierarchical multistage porous structures [2]. Multistage porous structures are widely found in nature, such as bamboo and loofahs, which possess reasonable pore size distributions and excellent surface properties, such as high surface strength [55], larger specific surface area, and more reasonable pore distribution [2,24]. These porous structures show great fascination in various fields [56]. As shown in Fig. S4 (Supporting information), the structural properties of different biomimetic porous materials and their applications in functional adsorption and mechanics are listed. In addition, researchers have found that porous structures tend to have strong energy absorption capabilities, contributing greatly to fields such as energy absorption and storage [57]. For example, based on the porous structure of butterfly wings and sea urchin skeletons, researchers combined 3D printing technology to bionically design a porous lightweight structure (Fig. S5 in Supporting information), which has an excellent energy absorption effect and at the same time has excellent surface strength. Moreover, the porous structure is heterogeneous, and the heterogeneous structure is extremely beneficial in promoting bone growth and blood vessel healing [58]. And these are only a small part of the role of bionic porous structure. Multi-stage porous structures have excellent transport efficiency, a reasonable pore size distribution, and a large specific surface area [59], and these advantages make multi-stage porous structures also have great potential for application in the field of water treatment.

Meanwhile, different types of pore sizes in multistage porous materials affect the properties and functions of the material surface [23]. Micropores play an important role in multistage porous materials due to their small pore size and are crucial in catalysis [60], adsorption [61,62] as well as membrane separation [63]. The selective action of micropores allows for the selective passage of substances of specific sizes as well as specific particles to improve the efficiency of the action of porous materials [21], such as the distribution of micropores within a plant, the effect on its transpiration, photosynthesis [64], and nutrient transport [65]. Mesoporous structures are widely found in animal bones and keratin, with a high specific surface area, which extends the range of action of micropores and shows great potential in adsorption and bone growth [21].

Widely found in various structures in nature, macropores promote particle transport due to their large pore size, which provides large spatial transport channels. In addition, macropores provide elasticity and structural support, and an excessive distribution of macropores may lead to the collapse of porous structures. Whether it is a macroporous, mesoporous, or microporous structure, a reasonable distribution of pore sizes plays an important role in the mass transfer and transport of pollutants in bionic multistage porous structures [21].

Therefore, natural multistage porous materials have a more reasonable structure in adsorption, which makes multistage porous materials have unique advantages in the adsorption of pollutants in wastewater, such as heavy metals, pigments, and oil [66]. Hence, mimicking and preparing natural multistage porous structures and applying them to water treatment adsorbent materials is important.

The following section provides an overview of the different existing biomimetic objects with porous structures in nature, together with a description of the characteristics of their pore sizes and their potential application in water treatment, in addition to describing the study of the application of such multistage porous structures in graphene water treatment adsorbent materials. Among them, Table S2 (Supporting information) lists the existing biomimetic multistage porous structures in nature and the fea-

sibility of their application in adsorbent materials for water treatment.

2.2.1. Porous structure of bamboo

The porous structure of bamboo plays an important role in bamboo growth, nutrient transport, and so on. The porous structure of bamboo is not only reflected in the bundle-like structure on the bamboo nodes but also in its tubular structure, as well as at the nodes; the same porous, cellular structure exists, as shown in Fig. 3a. These combined structures have a wide range of applications in fields such as energy-absorbing materials, especially in the adsorption of pollutants such as heavy metals, which have excellent potential. The tubular-like structure combined with the porous, cellular structure is more favorable for the adsorption, mass transfer, and storage of pollutants. At the same time, the unique porous structure of bamboo gives it large stiffness and mechanical strength. For example, it can withstand high bending stresses from gusty winds, and especially, the cylindrical structure makes bamboo have better compressibility than ordinary materials [67]. Such excellent mechanical properties are also important in water treatment adsorbent materials [68].

The multistage porous structure in biomaterials has the effect of improving material transport. Xue *et al.* [69] prepared mineralized calcium phosphate/bamboo composite scaffolds with excellent mass transfer properties and strength by biomimetic mineralization and biomodelling methods. Researchers have biomimetically prepared different kinds of adsorbent materials inspired by the porous structure of bamboo. Chen *et al.* [70] prepared bamboo charcoal adsorbents by activation with potassium permanganate and sulfuric acid, which greatly increased the specific surface area of bamboo charcoal, making it highly adsorbable for bromate (BrO_3^-).

The above studies have, to some extent, utilized the porous structure of bamboo for the excellent mass transfer properties it possesses. Bionic scaffold materials were prepared, which embodied its excellent transport properties. In addition, how to borrow the bamboo structure and apply it to graphene water treatment adsorbent materials has attracted extensive attention from researchers. For example, to solve the structural disorder in graphene aerogels and the disadvantage of hydrophobicity on its surface, He *et al.* [71] prepared pectin/graphene oxide aerogels (PTGAs) with bamboo-like structure by borrowing the structure of bamboo and using the template method, and the maximum adsorption amount of rhodamine B reached 719 mg/g in 80 min. The PTGAs also showed excellent cycling performance and stability performance. Excellent cycling performance as well as stability performance. To solve the problem of oil pollution of water resources. Huang *et al.* [72] prepared graphene aerogels with three-dimensional layered pores using bamboo and waste paper. Due to the tragic heterogeneity of bamboo, it provides well-developed pores for the graphene aerogel structure, which results in good adsorption properties (87–121 g/g) and elasticity.

2.2.2. Loofah

As a representative of nature's graded porous materials, Loofahs have excellent porosity and compressibility [73]. Its structure is widely used in the fields of energy storage materials and energy absorption materials [74]. The multistage porous structure of luffa contains a reasonable distribution of macropores, mesopores, and micropores with intricate pore diameters, forming a three-dimensional network structure (Fig. 3b), which makes the luffa itself have an excellent mass transfer capability [75]. Compared with other multistage porous materials, the loofah structure has the following advantages for application in adsorption: (1) The loofah itself has a complex structure and a large specific surface area. (2) Loofah has a porosity of up to 93%, with a reasonable pore size

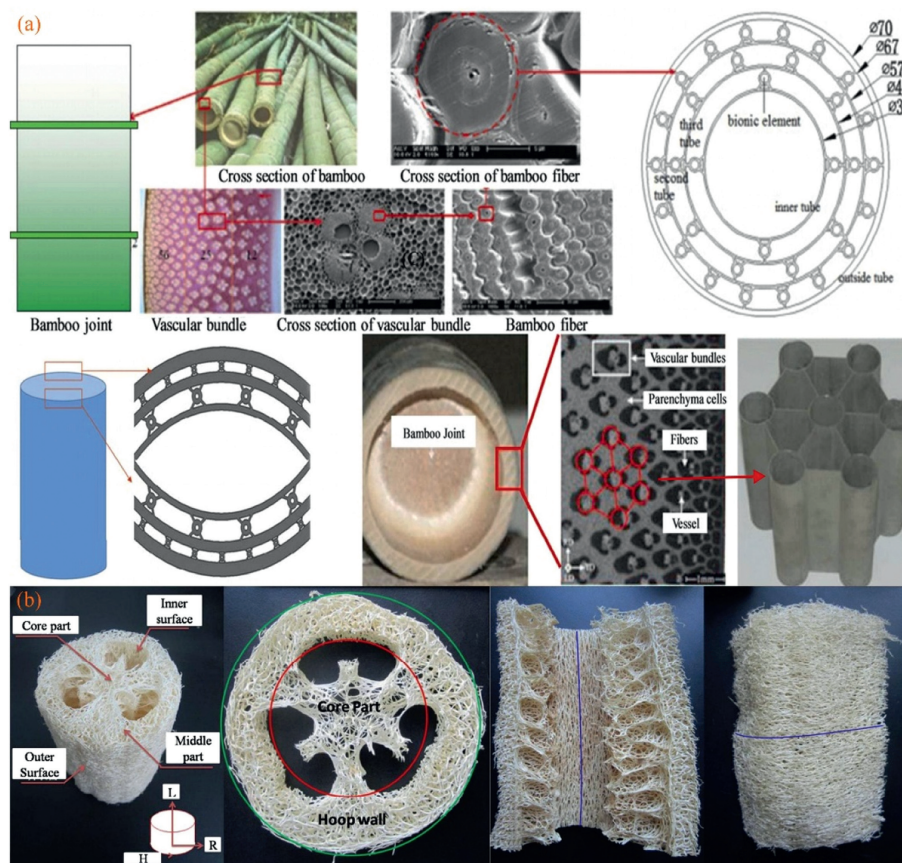


Fig. 3. (a) Porous structure of bamboo and its bionic model. Reproduced with permission [2]. Copyright 2022, Elsevier. (b) Porous structure of luffa and its different parts. Reproduced with permission [78]. Copyright 2014, Elsevier.

distribution and excellent transport properties. (3) Loofah has low flow resistance, as well as low density. (4) Loofah has high strength and wear resistance.

Researchers are committed to promoting the use of luffa in adsorption. For example, Li *et al.* [76] prepared excellent loofah biochar adsorbents from loofah by chemical modification of surface functional groups. The adsorption of uranium could reach 247.58 mg/g. The activation of the loofah sponge by granular materials improved its ability to adsorb fluoride ions from water. The adsorption experimental study found that the activation by surface hydroxyapatite resulted in the optimal performance of the adsorption of fluoride ions. It was pointed out that the oxygenated group (-OH) in the loofah structure is the main reason for the high adsorption performance. Khadir *et al.* [77] reviewed the removal of pollutants, such as heavy metals and dyes, by loofah structure and also explored the effect of adsorption time, temperature, and other factors on the adsorption process and finally compared the study of raw and modified loofah. It shows that loofah structure has good potential for application in water treatment.

The luffa porous structure and mechanical properties of lucerne offer potential advantages for water treatment applications [78]. Most of the above studies are limited to using loofah to make biochar adsorbents or the preparation of loofah-based adsorbents by surface doping and modification. There is a lack of direct utilization of the unique structure of the loofah and its application to adsorption cases. Surface doping modification makes the original structure of loofah unable to play an optimal effect and, at the same time, has certain defects. Currently, there are fewer studies on combining the loofah structure with graphene or applying the loofah structure mimicry into graphene, and it is a hot research topic in the future to focus on accurately mimicking the loofah

structure and applying it to graphene water treatment materials as well as to exert the treatment of pollutants by the loofah structure itself.

2.2.3. Corn cobs

Corn cobs have a porous hierarchical structure. At the structural level, it is divided into the pith, lignin rings, and glumes [79], where the lignin rings are the main source of mechanical strength, and the structure of the lignin rings is a hollow tubular structure composed of cellulose. The porous structure of the pith and glumes exhibits a porous sponge-like structure, possessing a large specific surface area and a well-developed pore structure [79]. The multicellular microstructure of the pith makes the corn kernel possess better compressive and cushioning properties, while the irregularly raised, elliptical structure of the glumes contributes a large specific surface area.

Corn cob has great potential in the field of adsorption. For example, Ismail *et al.* [80] prepared a corn cob adsorbent for the adsorption of malachite green dye by treating corn cobs with concentrated sulphuric acid. The adsorbent showed good adsorption properties for malachite green, especially at low concentrations of malachite green solution. This study provides a reference for the treatment of dye wastewater and the application of corn cob materials. Sciban *et al.* [81] investigated the adsorptive removal of heavy metal ions by different biomasses. In order to ensure the adsorption capacity and to prevent the leaching of pollutants, the corn cob material was treated by washing it with water. The results showed that the treated corn cob had excellent adsorption effect on cadmium and nickel ions. This study can make biomass adsorbents such as corn cob better used in water treatment adsorbent materials. In addition, Liu *et al.* [82] prepared activated car-

bon from corn cobs using KOH as an activator. It was found that the activated carbon achieved effective adsorption and removal of Hg(II). There are few studies on the bionic application of corn cob to graphene materials, and only An *et al.* [83] reported a bionic-functionalized corn cob/reduced graphene oxide aerogel (CL/rGO) prepared by freeze-drying technique, and the adsorption and cycling experiments showed that (CL/rGO) has good adsorption properties for dyes, as well as good recycling properties.

The pith and glumes in corn cobs have a well-developed porous network structure. This porous structure can be utilized directly or combined with modification to prepare adsorbent materials that perform excellently. However, direct replication of the corn cob structure for water treatment is less studied. The main reasons are as follows: (1) Although corn cob has a porous structure, the pore size is small, which is difficult to replicate through traditional techniques. (2) The printing accuracy of existing bionic methods, such as 3D printing, needs to be further improved.

The pith and glumes in corn cobs have a well-developed porous network structure. This porous structure can be utilized directly or combined with modification to prepare adsorbent materials that perform excellently. However, direct replication of the corn cob structure for water treatment is less studied. The main reasons are as follows: Although corn cob has a porous structure, the pore size is small, which is difficult to replicate through traditional techniques. In future research, it is necessary to work with some modeling software to tune its pores to the right size, and in conjunction with the AM technique, a complete replication of the structure would be an effective strategy.

2.2.4. Bone structure

Bone is divided into cortical and cancellous bone. In nature, cancellous bone is multistage and porous, with a large specific surface area and well-developed pores. Cancellous bone is widely found in living organisms such as cow bones and dog calf bones. Researchers have found that cancellous bone also has the heterogeneity of the Voronoi-Thiessen diagram [58], and it is the existence of this heterogeneity that makes cancellous bone better for nutrient exchanges and possesses great structural stability [84]. Researchers have applied it in different fields, such as bone tissue engineering, as shown in Fig. S6 (Supporting information) below, which demonstrates the porosity as well as the heterogeneity of cancellous bone and its application to novel bone tissue engineering.

Bone structure is a natural multistage porous structure that has great potential in the field of adsorption. Yang *et al.* [23] used bovine bone as a raw material, which was pyrolyzed at a high temperature of 450 °C and then surface modified with H₃PO₄ and activated with K₂CO₃. Adsorbents (VOCS) with excellent adsorption properties for VOC were prepared. The mass transfer performance of the adsorbent was excellent; the pore volume of the adsorbent reached 2.807 cm³/g, and its specific adsorption capacity of VOC was 13.03 mmol/g. The adsorption performance was much higher than that of toluene under the same conditions, and it had excellent regeneration performance.

The multistage porous structure of bone has the disadvantage of having too small pores, although it possesses a developed pore structure and a large specific surface area. Therefore, future research will focus on how to accurately replicate the porous channels of bone tissue and its structure into graphene and other adsorbent materials.

In summary, structures such as bamboo and loofah in nature have a well-developed porous structure. This structure plays a great role in nutrient transport and adsorption of pollutants in water. In addition, the porous structure has well-developed pores and excellent mass transfer performance, which can also provide a more reasonable pore distribution and a larger specific surface

area for graphene adsorption of pollutants. At the same time, a reasonable pore size distribution will also optimize the force structure of the macroscopic body [23], which can provide an optimal solution strategy for graphene facing strong hydraulic conditions, resulting in a great improvement of graphene adsorption performance and mechanical properties. This improvement is often difficult to achieve by traditional means, thus making the porous structure have great potential in the field of water treatment adsorption.

Whether it is function bionics or structure bionics, it has an important role and significance in improving the adsorption performance of materials, which can be completely applied to graphene structure. At present, function mimicry is mainly based on the related properties of lotus leaf and spider silk, etc., through surface modification to improve the adsorption performance of graphene. Or through the use of functionalization on the surface of graphene to mimic the superhydrophobicity of lotus leaves. And graphene materials with such superhydrophobicity can be effectively applied to the field of oil adsorption. Unlike mimicking functions to improve the adsorption performance of graphene, structural biomimicry improves the performance of graphene adsorption through the large specific surface area provided by the porous structure as well as the well-developed pore space (7), which provides fast pathways and a large adsorption space for the transport and storage of pollutants [22,23]. Table 1 [7,26,43,45,85,86] summarizes the current work available on graphene-based biomimetic adsorbent materials for water treatment and compares their performance. Nature provides great space for the biomimetic design of graphene water treatment adsorbent materials. As of now, the research on the biomimetic study of water treatment adsorbent materials including graphene is not perfect enough and there is no systematic description. Therefore it still needs to be explored continuously to provide a more perfect theoretical basis.

3. Preparation method of graphene-based biomimetic material

3.1. Biomimetic mineralization method

Biomimetic mineralization is an effective way to biomimetic preparation of bionic materials; this method is a simple way to precisely regulate inorganic substances through specific biomolecules so they can be assembled into minerals [87]. Compared to methods such as self-assembly, the biomimetic mineralization method has the advantage of controlling the preparation of materials of specific sizes and shapes. For example, Yin *et al.* [88], in order to construct a stable superhydrophobic coating, a bionic mineralization layer was introduced into the coating under the induction of dopamine, and the nanosilver was continuously reduced under the reducing effect of dopamine and mineralized particles increased the hydrophobic structure of the material. Finally, the superhydrophobic coating with bionic antimicrobial properties was obtained under silane modification.

Research in biomimetic mineralization in biomimetics mainly centers around preparing specific biomimetic materials based on inorganic minerals. However, there are fewer reports on combining graphene materials and biomimetic technologies. As shown in Fig. S2 (Supporting information), researchers have prepared bionic graphene microsomes that can be shaped by bionic mineralization [89]. Cheng *et al.* [90], to mimic hydroxyapatite's mineralization process during bone formation, their team reported a reduced graphene oxide-induced hydroxyapatite mineralization material modified by dopamine. This material promotes osteoblast growth and adhesion, making it useful as a bone scaffold for bone growth. Liu *et al.* [91] also reported gelatin-modified graphene oxide (GO-Gel) for hydroxyapatite biomimetic mineralization, which has good potential in bone surgery. In addition, Tang *et al.* [92]

Table 1

Research status of graphene-based adsorbent materials for water treatment.

Absorbent material	Bionic object	Removal of contaminants	Adsorption capacity	Removal efficiency	Ref.
Mt/GO/HA	Coral	Cu ²⁺	198.8 mg/g	88.30%	[7]
CNF/PEI@GOA	Spider silk	Cd(II), Cr(VI), Cu(II), Pb(II)		100%	[26]
LDH@PDA@MPNs	Mussel	Crystal violet (CV), Malachite green (MG)	89.608 mg/g (CV) 40.481 mg/g (MG)	98%	[43]
MWCNT-PDA-CS-GO	Dopamine	Gd(III)	150.86 mg/g		[45]
KGCA	Konjac glucomannan	U(VI)	513.4 mg/g		[85]
3DPLA@GO/Ccs	Fish mouth structure	Crystal violet (CV)	46.2 mg/g	97.80%	[86]

reported a three-dimensional mesh graphene/hydroxyapatite drug-carrying material prepared by biomimetic mineralization, in which hydroxyapatite was first modified with citric acid, followed by biomimetic mineralization to form an apatite layer on the hydroxyapatite/graphene surface. The composite material has a good loading effect on aspirin. Furthermore, Liu *et al.* [7] investigated the trapping role of corals by bionic mineralization combined with graphene oxide. GO biomimetic coral adsorbent material (Mt/GO/HA) with excellent sensing and trapping properties for metal ions was prepared. The material has a compact structure with uniform and well-developed channels exhibiting a large specific surface area while exposing a large number of functional groups. This enabled the bionic coral structure to rapidly adsorb Cu²⁺ with an adsorption capacity of 198.6 mg/g. It was shown that the mechanism of synergistic adsorption of heavy metal ions by hydroxyl (-OH) and carboxyl (-COOH) groups in Mt/GO/HA played a major role (e.g., Fig. 2b), and the bionic adsorbent material had a very good recycling performance.

Although the bionic mineralization method has a wide range of applications in the preparation of structural bionic and functional bionic materials, it is hampered by the limitations of bionic mineralized materials and the incompleteness of their bionics.

3.2. Template method

The stencil method has been effectively used in bionanomaterial preparation as a practical approach to synthesizing nanomaterials with a great deal of flexibility (e.g., Fig. S3a in Supporting information). Zou *et al.* [93] prepared nanomaterials bionic coral-loaded nanomaterials by loading graphene oxide on PANI using gaseous carbon formed from calcium carbonate salts as a stencil. The prepared materials have good corrosion resistance and self-healing properties. Weng *et al.* [5] prepared thin films of graphene oxide with an asymmetric surface structure using the template method to prepare optically driven actuators with complex shapes that are self-healing and enable programmability. This material study provides a reference for researching programmable and self-repairing light-driven robots. In addition, Zhou *et al.* [94] prepared hydrophobic nanofibre/poly(vinyl alcohol)/graphene oxide composite aerogel materials by using ice crystals as a stencil, which have not only high adsorption efficiency but also have good strength and good potential for application in oil adsorption. The template method can rely on preparing different stencils to create bionic materials with different structures and functions. However, due to the disadvantages that it is easy to destroy the material structure when removing the stencils and the tedious preparation process, it is challenging to prepare functional and structural biomimetic materials well.

3.3. 3D printing method

Although the above methods have many advantages in the preparation of graphene-based biomimetic materials, they face the disadvantages of not being able to accurately replicate the specified structure, having poor bionic performance, and not being able to

be integrally molded [95]. In contrast, 3D printing technology has the advantage that it can be prepared layer by layer, fully molded, and completely bionic (Fig. S3b in Supporting information) [96,97]. Table S3 (Supporting information) makes a comparison between the advantages and shortcomings of different bionic preparation methods.

Therefore, researchers are committed to the use of 3D printing technology in structural biomimetic materials, which is becoming an important tool for the preparation of biomimetic materials compared to other methods. For example, Peng *et al.* [98] used a new class of cellular bionic structure (TPMS), which exists in nature's butterfly wings and sea urchin skeleton structure, to prepare TPMS structure by 3D printing technology and analyze its properties. The results showed that TPMS has good mechanical properties and energy absorption ability. Yang *et al.* [99] prepared 3D printed material with superhydrophobicity inspired by the superhydrophobicity of plant leaves, which can be effectively used for oil pollution treatment.

In addition, the researchers also explored 3D printing technology in the study of graphene-based biomimetic materials.

Li *et al.* [100] prepared a biomimetic microstructure using the 3D printing method, which can be effectively used in biosensing. Liang *et al.* [101] reduced graphene oxide-based biomimetic cellular microstructures were prepared using rGO as a material using the 3D printing technique. Glass *et al.* [102] were inspired by the biological and used polycaprolactone and graphene as a material for the 3D printing of artificial cilium arrays. As for the study of graphene-based bionic water treatment adsorbent materials, Zhou *et al.* [86] reported a poly(lactic acid)/graphene oxide/chitosan filter with a bionic fish mouth structure. The filter exhibited excellent adsorption removal efficiency for crystalline violet (CV) of 97.8%, and showed great potential for the adsorption of organic dyes. In addition, Masud *et al.* [103] improved the rheological properties of graphene by adding two biomimetic polymers, PDA and bovine serum albumin (BSA), to the graphene ink. 3D graphene biopolymer aerogel (G-PDA-BSA) was 3D printed using direct ink writing (DIW) method. Adsorption experiments showed that G-PDA-BSA has excellent removal performance for heavy metal ions, organic dyes, organic dyes, *etc.*, in addition to the excellent regeneration potential of G-PDA-BSA.

Research on the fabrication of graphene-based bionic adsorbent materials for water treatment using 3D printing technology is currently evolving, with continuous attempts by researchers. However, the unique advantages of AM technology combined with bionics make 3D printing an excellent potential for the preparation of graphene water treatment bionic adsorbent materials. Whether in the preparation of graphene functional bionic materials or graphene structural bionic materials, 3D printing technology has the advantages of integrated molding and controllable structure; the only difference is that the former needs to carry out the expected modification of the ink before printing, while the latter needs to carry out the identification and differentiation treatment of the printed structure, and for the next step of the printing process, both are the same. This one-piece preparation method optimizes the preparation process of graphene bionic adsorbent mate-

rial, making it more controllable, and also avoids the shortcomings of other methods with poor bionic effect. It makes the prepared structure more precise and stable. It improves bionic efficiency. 3D printing technology has significant advantages for preparing graphene-based bionic adsorbent materials for water treatment.

4. Application study and prospect of 3D printed graphene biomimetic adsorbent

3D printing, also known as additive manufacturing, is an umbrella term for a class of technologies instead of "subtractive manufacturing" [104]. After more than 30 years of development, AM technology has been widely used in aerospace, medical, and other cutting-edge fields. As a practical and low-sinking cost tool, AM technology can be well applied to constructing bionic structures [105]. AM technology has a similar method to the synthetic class of the deposition of substances in the organism and has unparalleled advantages compared to traditional manufacturing methods [2]. AM technology can be used to build up the internal features of bionic structures step by step by printing the pore size and structure of each layer by programmed control (Fig. S7 in Supporting information). The complex internal structure of the bionic structure, using traditional methods can not accurately simulate its microstructure. AM technology can be accurately controlled, and layer-by-layer step-by-step printing [106] is a complete simulation of the bionic porous structure of the natural world. AM technology can also print materials for specific functional modifications to achieve the bionic function. All of these advantages of AM are unachievable by traditional methods.

Currently, graphene 3D printing technologies are available: inkjet printing [107], DIW [108], and fused deposition printing (FDM) [104]. Inkjet printing is a new contactless printing method, which generally involves spraying many ink droplets onto the substrate and then depositing the print to form. In addition, inkjet printing has higher requirements for the nozzle and ink viscosity, the printed items have lower structural accuracy, and the printing process needs to be dried, making the process more cumbersome and the printing success rate lower. FDM technology generally uses the rayon machine to prepare the material, and then by heating the material, the material will be sprayed from the nozzle by the programmed graphics and layer-by-layer stacking manufacturing. However, FDM technology relies too much on the material and extrudes the material too quickly to be precisely controlled; in addition, there are variations in the temperature of the print, making the printed material prone to collapse. DIW technology, on the other hand, is designed by a computer program to prepare 3D structures by controlling the movement of the nozzle and continuously extruding the slurry from the nozzle.

Compared to the previous two printing technologies, DIW technology is more widely used and requires less material and more accurate printing. Moreover, DIW technology has a wide range of applications and can effectively prepare a variety of complex structures [104]. Therefore, it is more advantageous to prepare graphene biomimetic materials by direct writing, and the DIW technology can be used to prepare better functional and structural biomimetic materials (Table S4 in Supporting information). Functional bionics is achieved more simply by adding additives, such as mussels, to the printed paste and preparing 3D graphene through integrated moulding. The preparation of structure bionics requires the use of scanners, etc., to convert the structure to be imitated into a computer language and then, under the control of the program, control the movement of the nozzle to reproduce the bionic structure.

However, this technology is currently less used in the field of graphene water treatment, so it needs to rely on related research in other fields. For example, the method of preparing new bionic materials by combining 3D printing technology and bionic struc-

tures has been developed in the preparation of energy-absorbing, lightweight, and surface multifunctional materials. For example, Chen *et al.* [109] prepared bionic loofah bone scaffolds of hydroxypapatite by using loofah as a bionic object, taking advantage of the multistage porous structure of loofah, which exhibits excellent surface properties in terms of osteoblast growth, as shown in Fig. S8 (Supporting information), which demonstrates the structure of bionic loofah, and the printed model of bionic scaffolds. The loofah has a porous reticular structure with a porosity ranging from 79% to 93% and a large specific surface area. The porous structure, along with the large attachment area, allows cells to attach and grow on its surface, making the porous structure great potential in the field of bone growth. Its excellent mass transfer properties, demonstrated by the multistage porous structure of luffa, allow for the good transfer of cellular nutrients. The excellent transport properties of the porous structure, as well as the high specific surface area, also have immeasurable potential for application in adsorbent materials for water treatment.

It is clear that the preparation of graphene-based biomimetic adsorbents using 3D printing technology has extremely excellent potential, but fewer studies have been reported. Only Zhou [86] and Masud [103] provided relevant research.

Overall, nature's specific structures and properties have great potential for adsorption, but so far, there have been very few studies combining 3D printing technology, bionic structures, and graphene materials. In addition, the research on the fabrication of graphene-based biomimetic adsorbents for water treatment using 3D printing technology also has some defects and difficulties. The main difficulties in preparing biomimetic adsorbent materials with graphene structure are: (1) It is difficult to accurately identify and replicate some tiny, porous structures, such as some complex nanoscale structures, with the existing technology. Moreover, these tiny structures play an essential role in the functioning of the adsorption process. (2) Most 3D printing manufacturing technologies have low precision. They cannot accurately reproduce the identified structures, which makes the prepared bionic structures differ from the expected ones and fail to play their proper effects. (3) Higher printing costs: although some industrial-grade 3D printers can achieve printing accuracy of about 20 μm , the printing cost is expensive and cannot be widely used. The main difficulties in 3D printing graphene functional bionic adsorbent materials are: (1) Prior to graphene functional biomimetic 3D printing, the graphene material needs to be functionally modified to achieve its specific functions. However, functional modification may cause the materials to wrap around each other and affect their performance. (2) The modified material often requires a specific matching printer to complete the printing, and this specificity often limits the preparation and application of graphene-functional bionic adsorbents. In addition, some common factors, such as the properties of printing materials, further limit the preparation of graphene-based bionic water treatment adsorbent materials by 3D printing. Regardless of the 3D printing preparation of functional or structure bionic graphene-based adsorbent materials, the printed materials must meet specific rheological properties, stability, and mechanical properties. Graphene material has the advantages of a large specific surface area and better biocompatibility than other printing materials. The defects of graphene materials in printing are poor rheological properties, after printing or freezing, prone to deformation, collapse, and other hazards, making the printed material pores or the overall structure of the deformation, and so on, often needing additional auxiliary conditions.

Although there are some difficulties in preparing graphene-based water treatment adsorbent materials by 3D printing at present, this makes research progress slower, and there are fewer cases of application in water treatment. However, the combination of 3D printing technology and biomimetic technology has

shown excellent results, which can further promote the development of water-treatment adsorbent materials, especially in applying graphene water-treatment adsorbent materials. The potential of graphene materials in water treatment can be further explored so that 3D graphene can better play its adsorption performance and better treat the pollutants in water. Future research focuses on:

- (1) How can we better develop and utilize the potential of 3D printing, improve the accuracy of the printers, refine the printing technology, and reduce costs.
- (2) Combined with computer technology, complete identification and replication of natural multilevel porous structure, combined with 3D printing technology, which is applied to graphene and other water treatment adsorbent materials, to improve the adsorption properties and mechanical properties of 3D graphene.
- (3) Improve the stability of the material to prevent the collapse and expansion of graphene while not affecting the original adsorption properties of graphene; it also prevents wrapping between modified materials and printing materials such as graphene.

5. Future developments

Currently, bionics techniques have attracted much attention from researchers in various fields, showing tremendous potential. Similarly, nature also provides unique ideas for the development of water treatment materials, and, compared with bionic methods such as template method and biomimetic mineralization method, 3D printing technology has the advantages of complete bionic, easy moulding, rapidity, and efficiency in one-piece preparing graphene-based biomimetic water treatment adsorbent materials. In this paper, the research and related progress of graphene-based bionic water treatment adsorbent materials are reviewed from the aspects of structural bionics and functional bionics, and an overview of the research on graphene bionic technology combined with 3D printing technology is also given. Although the research on the application of graphene-based bionic adsorbent materials for water treatment, especially the preparation of graphene bionic adsorbents by one-piece moulding using 3D printing technology, is still in the developmental stage, many research teams are continuously making breakthroughs. Based on the existing research, this paper provides some suggestions to face some gaps in the research of graphene water treatment bionic adsorbent materials and its combination with 3D printing technology and makes a prospect for the development of water treatment bionic materials, as follows:

- (1) Some unique features in nature, such as the lotus leaf effect and the amphiphilic nature of spider silk, have good bionic applicability in water treatment materials. By introducing these functions into water treatment adsorption materials such as graphene by bionic means, their adsorption performance can be effectively improved. However, the current research preparation process for graphene function bionics is partly polluted, an environmental safety hazard. In addition, in terms of the research on the preparation of graphene-based water treatment adsorbent materials using 3D printing technology, the modified printed materials, which are covered with each other, cause the effect of functional bionics to be unsatisfactory. This needs to be further improved and explored in future development.
- (2) The multistage porous structure is essential for graphene adsorption of pollutants because the existence of a multistage porous structure not only exhibits a large specific surface area but also has an essential significance for the transport of pollutants. However, most of the current research on multistage porous structures focuses on structural design

and energy absorption, and very few studies have been reported in the field of adsorption for water treatment. Due to the potential of 3D printing technology, this structure could be reproduced in its entirety by 3D printing and replicated in graphene structures in future research. However, at the same time, the graphene material should also be modified to prevent the swelling of graphene and collapse, which can cause the harm of the deformation of the bionic structure.

- (3) 3D printing technology has the advantages of integrated moulding and structural integrity, but it mainly faces defects such as insufficient printing accuracy, inability to print complex tiny structures completely, and poor material properties. Therefore, the following defects need to be solved to effectively use 3D printing technology and combine it with graphene water treatment adsorption material.

- (I) Improve the printer's accuracy, which can accurately print nano- and micrometre-scale structures.

- (II) To improve the relevant supporting technologies so that the functions and structures of the required bionics can be completely converted into computer language and the printer's operation can be controlled to achieve integrated printing.

- (III) Improve the mechanical and rheological properties of graphene materials to solve the problem of their tendency to collapse, swell, and deform during the printing process, resulting in structural deformation.

6. Summary

Graphene material has the advantages of a large specific surface area and high adsorption performance. However, the limitations of its own structure and character make it difficult to further improve its adsorption performance. Inspired by some plants and animals in nature, it is found that some of their excellent character and structures can be mimicked and applied to graphene materials so that the relevant properties of graphene can be further improved. These bionics can be broadly categorized into functional bionics and structural bionics. This paper begins with an overview of the different functional bionic and structural bionic objects existing in nature, introduces the relevant properties and advantages, describes the advantages and feasibility of their application in water treatment, as well as describes existing research related to graphene-based bionic adsorbent materials for water treatment. Secondly, several different existing methods for bionic preparation are described and compared. Finally, the advantages, as well as the feasibility of the application of additive manufacturing technology, represented by 3D printing, in graphene-based biomimetic water treatment materials, are described, as well as an overview of currently available 3D printing bionic technologies that can be drawn upon. Different existing 3D printing technologies and the flaws and shortcomings faced by 3D printing graphene-based bionic material are also compared and discussed. Nature has brought great inspiration to the development of water treatment materials, but how to effectively use the inspiration of nature and apply it to water treatment materials, as well as to clarify its inner mechanism of action still requires more in-depth research.

This paper hopes to provide reference and reference for further research development and application of water treatment bionic adsorbent materials. At the same time, it is expected to provide ideas for applying AM technology in biomimetic materials for water treatment so that the water treatment adsorbent materials can be better applied in the water treatment field.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

CRediT authorship contribution statement

Huining Zhang: Project administration, Funding acquisition, Conceptualization. **Baixiang Wang:** Writing – original draft. **Jianping Han:** Conceptualization. **Shaofeng Wang:** Investigation. **Xingmao Liu:** Investigation. **Wenhui Niu:** Investigation. **Zhongyu Shi:** Investigation. **Zhiqiang Wei:** Investigation. **Zhiguo Wu:** Investigation. **Ying Zhu:** Investigation. **Qi Guo:** Investigation.

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Supplementary materials

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