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Applications of flexible electrochemical electrodes in wastewater treatment: A review

Jiqing Liu[a], Qi Dang[a,*], Liting Wang[a], Dejin Wang[b,*], Liang Tang[a,b,*]

[a] *Key Laboratory of Organic Compound Pollution Control Engineering (MOE), School of Environmental and Chemical Engineering, Shanghai University, Shanghai 200444, China*
[b] *School of Resources and Environment, Anqing Normal University, Anqing 246011, China*

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ABSTRACT

The advancement of energy storage technology has paved the way for the application of electrochemical processes in achieving low-carbon and precise environmental pollution reduction. Electrodes play a crucial role in efficiently removing organic pollutants and heavy metals. To implement electrochemical pollution control technology in practical engineering, flexible electrode preparation is vital. This review highlights recent progress in flexible electrode research, focusing on the selection and structural design of flexible electrode materials. It summarizes the latest advancements in current collectors, active materials, and preparation methods to enhance conductivity, flexibility, and cycle stability. The application of flexible electrodes in water pollution control is categorized into three aspects: Organic pollutants, inorganic pollutants, and composite pollutants. Finally, the challenges and research requirements for enhancing electrode flexibility in environmental governance are discussed, along with prospects for their future applications.

1. Introduction

The presence of compound pollutants, including emerging contaminants like drugs and personal care products, has exacerbated water pollution issues [1–3]. Inadequate treatment methods result in a significant influx of polluted water into surface water or groundwater, posing a serious threat to the environment and human health [4,5]. Electrochemical technology has emerged as a promising and environmentally friendly approach to wastewater treatment, attracting significant attention due to its versatility, adaptability, and low-carbon characteristics [6].

Electrochemical technology utilizes the principles of electrochemistry and surface catalysis to remove compound pollutants, including organic pollutants and heavy metals. Through direct or indirect redox processes, electrochemical catalysis generates active species that effectively oxidize and degrade organic pollutants [7,8]. The electrode plays a vital role in the electrochemical reaction process, as its structural composition and active materials directly impact the efficiency of redox reactions, reaction mechanisms, and the extent of pollutant mineralization. Enhancing the electrochemical activity of electrodes is essential for improving the performance of pollution treatment systems [9–11].

Incorporating flexibility into electrode design is a strategy that addresses issues related to low efficiency and enhances the applicability of these systems in engineering contexts [12]. Flexible electrodes are functional materials that possess both electrical conductivity and the ability to undergo elastic deformation, enabling stable energy transfer even under conditions of deformation. They can be designed in various forms, allowing for customization based on parameters such as deformation degree, sensitivity, and energy density [13,14]. Flexible electrodes find applications in various fields, including medical sensing, energy conversion, aerospace, and wastewater treatment.

The introduction of flexible electrodes in wastewater treatment provides several benefits for advancing and improving the technology (Fig. 1):

(1) Enhanced contact and surface area: Flexible electrodes enable better contact and increased surface area with wastewater, facilitating improved electrochemical reactions and pollutant removal efficiency. The ability of flexible electrodes to conform to irregular shapes and contours enhances their contact with wastewater, ensuring a more comprehensive treatment process [15].

(2) Versatility in design and configuration: Flexible electrodes can be designed and configured in various forms to suit different wastewater treatment scenarios. They can be shaped into

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* Corresponding authors.
E-mail addresses: dangqi@shu.edu.cn (Q. Dang), wangdj@aqnu.edu.cn (D. Wang), tang1liang@shu.edu.cn (L. Tang).

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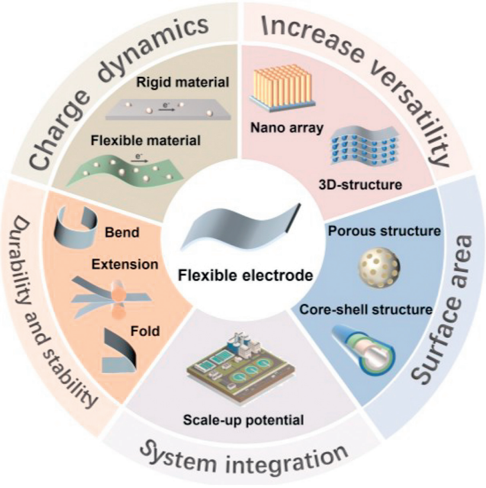

Fig. 1. Advantages of flexible electrode in wastewater treatment.

membranes, films, fibers, or other tailored structures, allowing for efficient integration into existing treatment systems or the development of novel approaches [16–19].

(3) Enhanced charge mass transfer and flow dynamics: The flexibility of electrodes allows for improved charge mass transfer and flow dynamics within the wastewater treatment system. The porous structure, three-dimensional structure and nanoarray structure can promote the heterogeneous interface diffusion between the electrode and the electrolyte, significantly shorten the electron transfer pathway and expose more active sites, thereby enhancing the movement and mixing of pollutants, ions and reactants [20,21]. This promotes more effective transport of contaminants to the electrode surface, increasing the overall treatment efficiency.

(4) Improved durability and stability: Flexible electrodes are often constructed using materials that exhibit excellent mechanical strength, corrosion resistance, and stability. These properties ensure the electrodes' long-term performance and reliability, even in harsh wastewater treatment conditions. Compared to rigid electrodes, flexible electrodes are less prone to mechanical failure, cracking, or delamination, resulting in extended operational lifetimes and reduced maintenance requirements [22].

(5) Scale-up potential and system integration: The flexibility of electrodes offers the potential for easy scale-up and integration into large-scale wastewater treatment systems. The adaptability of flexible electrode materials allows for their utilization in different reactor configurations, such as fixed-bed, fluidized-bed, or membrane-based systems [23]. This scalability enables the implementation of flexible electrodes in both centralized and decentralized wastewater treatment facilities.

Overall, flexible electrodes offer significant benefits in advancing wastewater treatment technology, facilitating improved pollutant removal and promoting cleaner and more sustainable water treatment approaches.

2. Fundamentals of flexible electrodes

Traditional electrodes have disadvantages such as inability to bend and poor stability, and cannot meet the urgent needs of flexible electronic products for high flexibility and machinability. Therefore, how to construct flexible energy storage devices is a research hotspot and difficulty. Flexible electrode is a kind of functional material with conductivity and elastic deformation ability. It is usually composed of flexible materials and inorganic materials with catalytic properties [24,25]. The composition and economy of flexible electrode be emphasized in Sections 2.1 and 2.2 in Supporting information.

3. Modified target for flexible electrodes

3.1. Electrical conductivity

The conductivity of flexible electrodes is a crucial factor that affects their performance. It depends on the conductivity of the conductive material used and the connectivity of the conductive path within the electrode [26,27]. To ensure stable electrical properties even after deformation or bending, several considerations should be taken into account when designing flexible electrodes:

(1) Improving the conductivity of conductive materials. High-quality conductive materials, such as high-purity metals or highly crystallized carbon materials (*e.g.* carbon fiber [28,29], graphene [30]), can significantly enhance the conductivity of flexible electrodes. Using these materials helps to ensure efficient charge transfer within the electrode.

(2) Selecting the appropriate thickness of the conductive material. The thickness of the conductive material is closely linked to the electrode's conductivity. Thinner conductive materials tend to have lower internal stress and provide better connectivity for the conductive paths, resulting in higher conductivity. However, it is important to strike a balance because extremely thin materials may compromise mechanical durability [31].

(3) Using a composite structure. Applying a protective layer on the surface of the conductive material can enhance its mechanical toughness and prevent fracture during deformation. This composite structure helps to maintain the conductivity of the electrode even under strain or bending [32].

(4) Optimizing the preparation process. The choice of an appropriate preparation process can improve the quality of the conductive material, thereby enhancing its conductivity. Different materials may require specific fabrication techniques to achieve the desired conductivity and mechanical properties [33].

More details about the methods for the electrical conductivity control of the flexible electrode are described in Sections 3.1.1, 3.1.2 and 3.1.3 in Supporting information.

3.2. Flexibility

Flexible electrode flexibility refers to the ability of the electrode to maintain electrical conductivity under various mechanical deformation conditions such as bending, stretching, and torsion, without deformation, rupture, or performance degradation. In the field of materials, the preparation of more flexible and even stretchable materials is a research hotspot [34]. The development of flexible electrodes that can withstand mechanical tensile deformation and maintain excellent mechanical properties and electrochemical properties. The preparation of flexible electrodes needs to explore the method of compounding the active material with the flexible substrate, and strengthen the combination of the two to improve the performance of the flexible electrode.

3.2.1. Material selection

Flexible current collector materials usually have high ductility, tensile strength and low brittleness, which can adapt to different shapes and sizes. In order to prepare flexible electrodes, electrode active materials and conductive additives are usually added to flexible substrates. Common electrode active materials include metal oxides, polymer nanoparticles, *etc.* While conductive additives can be conductive polymers, carbon nanotubes, conductive fibers, *etc.* Through the introduction of these additives, the conductivity and mechanical strength of the flexible electrode can be improved, thereby ensuring its electrochemical performance under complex deformation.

Conductive polymers are a class of materials with high conductivity and effective electrochemical energy storage characteristics, which have been widely used in the field of flexible electrodes. In order to apply conductive polymers to the preparation of flexible electrodes, conductive polymers can be directly deposited on the surface of flexible polymer substrates by electrodeposition. Chen *et al.* [35] prepared CNFs by electrospinning polyacrylonitrile nanofiber network carbonization, then electrochemically deposited polypyrrole, and then prepared a core-double shell structure CNFs@PPy@rGO composite electrode with a reduced graphene oxide coating. The rGO sheet is uniformly wrapped on the surface of CNFs@PPy to form a rGO film. The composite electrode maintains almost the same electrochemical performance at different bending angles, showing excellent flexibility. Polymer materials can also be made into films, showing good elasticity and tensile recovery, and can be stretched and reused. Fan [36] uses Ti_3C_2 MXene supported by cotton cloth as the electrode layer, and the cotton cloth is immersed in a mixed suspension of Ti_3C_2 MXene and cellulose nanofibers (CNFs). In the electrode layer, Ti_3C_2 MXene provides conductivity and electronegativity. The fabric substrate provides the strength and flexibility of the electrode layer. Zheng [37] prepared a sandwich-like carbon nanotube/$NiCo_2O_4$ composite paper electrode. Honeycomb-like $NiCo_2O_4$ nanosheets were coated on the surface of conductive carbon nanotubes, which combined the high conductivity of carbon nanotubes and the vertical arrangement structure of $NiCo_2O_4$ nanosheets. The CV curve of the composite paper electrode still has high electrochemical stability under mechanical deformation of 0° to 180°, and maintains 95.6% of its initial capacitance in 2000 cycles, indicating good cycle performance and flexibility. Yanilimaz *et al.* [38] prepared flexible polyaniline-carbon nanofibers (PANI-CNF) composites by preparing polyaniline coating on the surface of flexible carbon nanofibers to further improve the electrochemical performance of the electrode. Mechanical tests show that the independent PANI-CNF electrode has durability and high flexibility.

3.2.2. Preparation method

The preparation of different morphologies can also regulate the flexibility of the electrode. The introduction of fiber structures in electrode materials, such as nanofibers, nanowires or fiber reinforced materials, can increase the flexibility and tensile strength of the electrode, making it more resistant to bending and torsional deformation [39]. Choi *et al.* [40] embedded silver nanowires/reduced graphene oxide (AgNWs/rGO) electrode wires into the polyurethane (PU) dielectric layer on the PDMS substrate, and directly fabricated thin film electrodes on the elastomer substrate to prevent the change of electrochemical performance caused by stretching. The designed electrode is in the form of a network structure, similar to a honeycomb arrangement. Such a structure can make the electrode more flexible and bendable during bending and torsion, and reduce stress concentration. Kim *et al.* [41]. coated silver nanowires (AgNWs) on 2,2,6,6-tetramethylpiperidine-1-oxy-oxidized CNF nanopaper (TEMPO-CNF) to obtain AgNW@TEMPO-CNF substrate, and deposited mesoporous gold as a conductive electrode. Form dense anisotropic nano-scale creases and channel networks. The electrical signal remains stable under a strain bending of nearly 180°

In order to improve the overall flexibility, a multi-layer structure is designed to alternately stack different materials between the upper and lower layers of the electrode. Each layer of materials can complement each other to increase the flexibility of the electrode. Yin *et al.* [42] successfully prepared a layered core-shell SiCNWs@$NiCo_2O_4$NAS on a carbon cloth (CC) substrate using a SiC nanowire (SiCNWS) network as a nanoscale conductive skeleton. SiCNWs are intertwined to form a nano-scale SiCNWs network, which can not only serve as a nano-skeleton to support active materials, but also construct interconnected conductive electron transfer channels, and maintain its electrochemical performance at different bending angles.

3.2.3. Structural modification

Carbon nanotubes (CNTs) are nanomaterials composed of carbon atoms arranged in a tubular structure, which endows carbon nanotubes with high flexibility. CNTs also have excellent electrical conductivity, thermal stability, light weight, chemical stability, simple preparation and other advantages, so it has a good application prospect in the field of flexible electrode materials [43]. Zeng *et al.* [44] prepared carbon nanotube arrays on flexible carbon cloth (CC) by chemical vapor deposition. The Zn/CNT electrode has no obvious capacity loss (>97% retention rate) under different deformation conditions, indicating that it has excellent flexibility and mechanical strength. However, carbon nanotubes are prone to bending and winding, and agglomeration occurs, which affects their electrochemical performance [45]. Doping can change the electronic structure and surface properties of carbon nanotubes, thereby adjusting their conductivity, catalytic activity and nanostructures [46–48]. The doping method can be the hybridization of nitrogen, sulfur, oxygen and other elements, or the introduction of metal materials, such as copper and silver. Jiang *et al.* [49] deposited nickel nanoparticles on carbon nanotubes to prepare nickel nanoparticles@carbon nanotubes (Ni@CNT) composite electrodes, which made no binder between Ni and CNT and improved the performance of electrode materials. Flexible asymmetric supercapacitor (FASC) assembled with Ni@CNT as positive and negative electrodes has high volumetric energy and power density, good rate capability, excellent mechanical stability and long cycle life. Chen *et al.* [50] embedded Sb nanosheets into flexible porous N-doped carbon nanofibers (Sb@PCNFs) by a simple electrospinning deposition method. The Sb@PCNFs independent film formed by one-dimensional nanofibers has high flexibility and porous structure, which provides space for the volume change during charge and discharge and enhances the ion/electron transfer kinetics. In addition, the optimization and modification of carbon nanotubes can be achieved by adjusting the size and morphology of carbon nanotubes [51], and modifying functional groups on the surface of nanotubes [52]. Liu [53] prepared a high-performance self-supporting polyaniline (PANI)/single-walled carbon nanotube (SWCNT) film integrated electrode by a simple solution deposition method. Due to the strong π-π interaction between PANI and CNTs, a highly ordered interface layer is formed on the surface of CNTs, thus forming an efficient conductive network, which is conducive to the ion diffusion process and the rapid redox reaction at the electrode/electrolyte interface, so as to obtain better conductivity and enhanced capacitance performance. Through the comprehensive application of these methods, carbon nanotube flexible electrode materials with better performance and reliability can be prepared.

MXene is a typical two-dimensional metal carbide material, which has broad application prospects in the fields of electrocatalysis, energy storage and so on [54]. Among them, the stacking effect of MXene refers to the changes in the properties and structures of the composites composed of multiple MXene layers, which will affect the mechanical properties and thermal stability of the materials [55]. Therefore, it is necessary to further study its mechanism and characteristics to achieve effective regulation and utilization of MXene stacking effect. Liu [29] introduced a green, low-temperature-resistant and highly ionically conductive methane sulfonic acid/polyvinyl acetate (MSA/PVA) hydrogel as an electrolyte to optimize the performance of MXene, and used Ti foil as a current collector to form a flexible electrode. MSA/MSA ions are

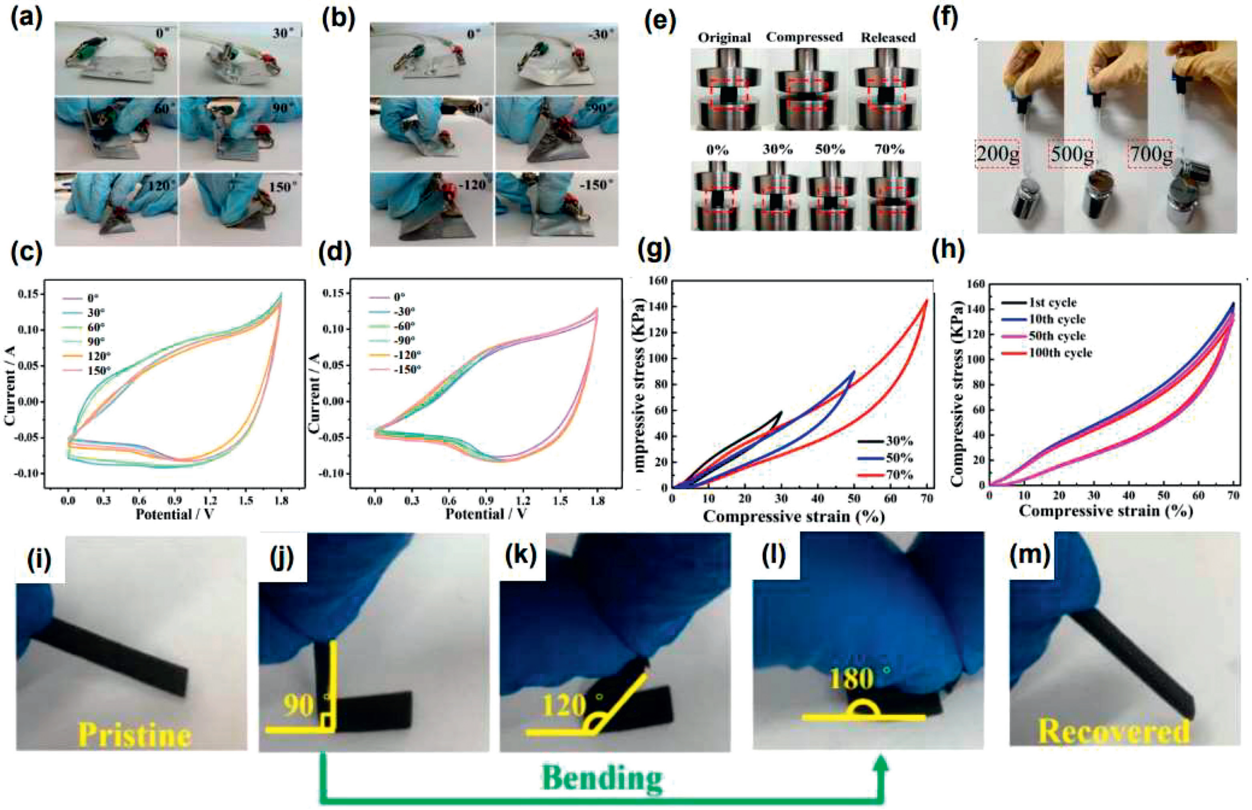

Fig. 2. (a, b) Digital photos of flexible equipment. (c, d) CV curves at different bending angles. Reproduced with permission [59]. Copyright 2023, Elsevier. (e) The digital images showing the compressibility of the MF/PPy-3. (f) The digital photos of tensile strength test for the MF/PPy-3/H_3PO_4/PVA composite using different weights. (g) The stress-strain curves of the MF/PPy-3 at different strains of 30%, 50% and 70%, respectively. (h) The stress-strain curves of the MF/PPy-3 at the strain of 70% for 100 cycles. Reproduced with permission [60]. Copyright 2019, Elsevier. Photographs of flexibility test using CSNS/NCF-160: (i) pristine, (j) under pressure, (k) pressure releasing, (l) after pressure released. (m) recovered. Reproduced with permission [61]. Copyright 2020, Elsevier.

pre-embedded in MXene, resulting in wrinkles and porous structures, thereby reducing the stacking effect of MXene and improving the flexibility of the electrode.

The three-dimensional structure flexible electrode has a multilayer and multi-stage structure, which can be designed and combined according to different requirements to form electrodes with different flexible properties [56,57]. The three-dimensional flexible electrode can maintain a high level of mechanical stability under various strain conditions such as bending, stretching and twisting [58]. Wang [59] prepared several $CuCo_2O_4$@NiCoS composite electrodes by controllable hydrothermal and electrodeposition routes. One-dimensional nanowires can shorten the transmission path of ions, while two-dimensional nanosheets expose many active sites. This makes the three-dimensional structure composite material have high electrochemical activity. It still has excellent mechanical stability after repeated folding at different angles (Figs. 2a-d). Sun *et al.* [60] prepared melamine foam/polypyrrole (MF/PPy) electrode with high mechanical properties and excellent electrochemical properties by *in-situ* interfacial polymerization of polypyrrole on commercial melamine foam. The three-dimensional cross-linked network structure makes the MF/PPy electrode material have excellent compressive deformation resistance and pressure reversibility. After compression, it can be completely restored to the original size and shape without plastic deformation (Figs. 2e-g). After 10 cycles, the compression curve remains basically unchanged (Fig. 2h). Ultrathin cobalt selenide (CoSe) nanosheets were successfully assembled on a three-dimensional nitrogen–doped carbon foam (CSNS/NCF) by Suo [61]. The foam is bridged by Co-C and Co-N-C bonds. CSNS/NCF not only has a large surface area in full contact with the electrolyte, but also provides a strong network with strong chemical bonds. The obtained CSNS/NCF-160 was bent to different angles (Figs. 2i-l). Obviously, CSNS/NCF-160 can be completely restored to the original shape after bending (Fig. 2m).

Therefore, through reasonable design and combination, flexible electrodes with better performance can be prepared, which is an important research direction for the development of flexible electrodes in the future.

3.3. Stability

Stability is one of the key indicators of the electrical properties of flexible electrodes, which is related to the composition and structure of the material itself. The flexible electrode with good performance should exhibit low resistance at the maximum working strain and remain almost unchanged during the cycle. It is difficult for the flexible electrode to maintain the absolute stability of the resistance during the deformation process, and the conductive material will inevitably undergo microstructure fracture during deformation recovery. The stability of flexible electrode is one of the important characteristics to ensure that the electrode maintains high conductivity during deformation or use. Because in the practical application of flexible electrodes, due to the deformation process such as bending, stretching or compression of the object, the conductive material will undergo microstructural fracture, which may lead to a decrease in the electrical properties of the electrode. Therefore, when designing and manufacturing flexible electrodes, it is necessary to consider their stability and take corresponding measures to improve their fracture resistance. More details about the material selection, preparation method and modification methods for the stability control of the flexible electrode are described in Sections 3.3.1, 3.3.2 and 3.3.3 in Supporting information.

4. Application of flexible electrode in wastewater treatment

Flexible electrodes have various applications in wastewater treatment, particularly in the removal of organic pollutants. The advantages of flexible electrodes, such as higher reaction rates, lower energy consumption, and environmental benefits, make them suitable for wastewater treatment processes. The treatment typically involves two steps: Anodic oxidation and cathodic reduction. Compared with the traditional electrochemical method, the flexible electrode has the advantages of flexibility and tunability. It can adjust or optimize its structure and function according to different pollutant types and water quality characteristics, and the effect is more obvious in practical application.

4.1. Organic pollutants

Traditional organic pollutants have been extensively studied and understood, and have a certain history in the environment, such as petroleum, polycyclic aromatic hydrocarbons, chlorinated organic compounds. Emerging organic pollutants refer to organic compounds that have emerged in recent years with less use and unclear environmental effects. Including drugs, personal care products, plastic additives, *etc.* These organic pollutants mainly come from the discharge of daily life and chemical production process. At present, the behavior and influence in the environment still need to be further studied and understood.

4.1.1. Conventional organic pollutant

Dyes are widely used in textile, leather, papermaking, food and beverage, medicine, cosmetics and other fields. Organic dye wastewater is a difficult problem in industrial wastewater treatment and environmental pollution control due to its complex composition, high toxicity and large emission. Jian *et al.* used diamond/carbon fiber with fiber core/shell structure as electrode, and the removal rate reached 89.25% within 100 min after adding 100 mg/L methyl orange solution [62]. Methylene blue (MB) is widely used in the fields of chemical indicators, dyes, biological dyes and drugs. Li *et al.* used a porous BDD/Ta film as an electrode to perform an electro-Fenton reaction at a current density of 60 mA/cm^2. After 80 min, the degradation rate of MB was 97% [63]. Azo dyes are the most commercialized dyes so far, accounting for about 70% of the total dyes. Their strong resistance to traditional processing and persistence in the environment pose environmental damage and health risks to organisms [6]. Acid Red G (ARG) is a water-soluble anionic azo dye with strong acidity. After 180 min treatment, the degradation rate of ARG on PbO_2/Pb_3O_4 electrode was more than 95.0% [64]. Alizarin yellow R (AYR) wastewater is a representative azo dye wastewater with high chromaticity, rich organic matter content and serious toxicity. After electrolysis of 80 mg/L AYR wastewater for 150 min, the AYR degradation rate and chemical oxygen demand (COD) removal rate of Ti/PbO_2-Sm_2O_3 composite electrode were 80.00% and 79.90%, respectively, and it still had excellent stability and catalytic activity after 10 cycles [65].

Naphthenic acid-rich aliphatics and their alicyclic carboxylic acids in refinery oil sand wastewater (OSPW) have attracted much attention due to their persistence, corrosion and toxicity. Abdalrhman used graphite anodes to treat dehydrated industrial naphthenic acid (NAs) mixtures in water samples at relatively low current densities. At current densities of 0.5, 2.5 and 5 mA/cm^2, the degradation rates of NAs were 76.9%, 77.6% and 82.4%, respectively, and the energy consumption was 0.086, 0.334 and 0.362 kWh/g NA, respectively [66].

Transition metal oxides and hydroxides are one of the most common electrode active substances, including RuO_2, MnO_2, NiO, SnO_2, Co_3O_4, NiCoO, $NiMoO_4$, $ZnCo_2O_4$, $Co(OH)_2$, $Ni(OH)_2$ and so on. The current research is mainly to prepare oxide materials with various morphologies, reasonable pore size distribution, large specific surface area and different lattice states on the basis of existing such materials. At present, there have been a large number of studies on the preparation and compounding of metal oxides and hydroxides, which can obtain different morphologies and properties.

Urea oxidation reaction (UOR) is considered to be a promising technology for simultaneous hydrogen production and degradation of urea-rich wastewater. The urea electrolysis reaction in alkaline environment can be expressed as (Eqs. 1-3) [67]:

$$\text{Anode}: CO(NH_2)_2 + 6OH^- \rightarrow N_2 + 5H_2O + CO_2 + 6e^- \quad (1)$$

$$\text{Cathode}: 2H_2O + 2e^- \rightarrow H_2 + 2OH^- \quad (2)$$

$$\text{Overall}: CO(NH_2)_2 + H_2O \rightarrow N_2 + 3H_2 + CO_2 \quad (3)$$

Sha [68] synthesized a self-supporting three-layer interconnected heterostructure $MnCo_2O_{4.5}$@Ni $(OH)_2$/NF, which is composed of Mn $Co_2O_{4.5}$ nanosheets seamlessly sandwiched between the bottom layer of the nickel foam substrate and the top layer of $Ni(OH)_2$ nanosheets. Without the use of binder, the active material is firmly bonded to the conductive substrate, which effectively improves the loading capacity of the active material and the flexibility of the electrode. The synergistic effect of $MnCo_2O_{4.5}$ and $Ni(OH)_2$ promoted the electrooxidation of urea. The key compound of nickel-based catalyst is NiOOH, which can be formed by the reversible electrochemical reaction of Ni^{2+}/Ni^{3+}. The corresponding electrochemical chemical mechanism is as follows (Eqs. 4 and 5):

$$6Ni(OH)_2(s) + 6OH^- \rightarrow 6NiOOH(s) + 6H_2O(l) + 6e^- \quad (4)$$

$$\begin{aligned} &6NiOOH(s) + CO(NH_2)_2(aq) + H_2O(l) \\ &\rightarrow 6Ni(OH)_2(s) + N_2(g) + CO_2(g) \end{aligned} \quad (5)$$

4.1.2. Emerging organic pollutants

Pharmaceuticals and personal care products (PPCPs) are a unique emerging environmental pollutant. PPCPS is becoming a widespread pollutant in aquatic and marine environments due to its wide application range, poor biological metabolism and unreasonable disposal. At the same time, it can also produce endocrine disruption, neurotoxicity and developmental toxicity, which have a negative impact on ecosystems and human health [69]. Liu *et al.* used Fe_3O_4NP@CN flexible electrode to almost completely degrade carbamazepine after 3 h of reaction, and the energy consumption was about 0.239 kWh/g carbamazepine [70]. Tin dioxide (SnO_2) is an N-type semiconductor with chemical stability and corrosion resistance. Titanium-based SnO_2 electrode has attracted much attention due to its high oxygen evolution potential, good stability and high catalytic activity. SnO_2 doped metal elements can improve the conductivity and oxygen evolution overpotential of the electrode. Due to the difference in the atomic radius between SnO_2 and doped metal, the electrode will produce chemical bond structure and crystal defects, thereby improving the catalytic strength and current efficiency. Duan synthesized copper-doped Ti/SnO_2 electrode by thermal decomposition method. The metal coating of Ti/SnO_2-Cu electrode is evenly distributed, and a highly rough, porous and well-layered active layer is obtained. A three-electrode system was constructed with Ti/SnO_2-Cu electrode as the working electrode. The degradation rate was 97.5% after 60 min reaction at 20 mA current [71]. Duan *et al.* used the G/CNT-Ce/PbO_2-Ce composite electrode for electrocatalytic degradation of ceftazidime. After 120 min of reaction at a current of 40 mA, the removal rates of ceftazidime and TOC reached 100.0% and 54.2%, respectively [72]. Zhou used a porous Ti/SnO_2-Sb anode to electrochemically degrade 5 mg/L abacavir. At a current density of 0.2 mA/cm^2, the degradation efficiency exceeded 97% within 10 min, the degradation rate constant was 0.36 min^{-1}, and the lowest energy of each order was 6.5 mWh/L [73]. Tetracycline is a kind of antibiotic widely used in clinical treatment. Because of its difficult biodegradation, it is easy to enter the soil with animal feces and eventually enter the surface water body, seriously affecting the ecosystem. Tetracycline was degraded by Fe/Co-CNFs electrode, and the degradation rate was close to 100%. The electrocatalytic degradation process has a high Faraday efficiency of 81.29% and a low power consumption of 6.17 kWh/kg [23]. When Ti/Ti_4O_7 was used as the electrode, the degradation rate of tetracycline reached 95.8% [74]. Dong *et al.* used CF@PANI@MIL-101 (400) as the cathode, and the initial concentration of *p*-nitrophenol and TOC were both 50 mg/L. After 120 min electrolysis at a current density of 5 mA/cm^2, the removal rate of *p*-nitrophenol was about 100%, and the removal rate of TOC was 52%. After ten cycles, about 94% of *p*-nitrophenol can still be removed [75]. Lignin is a kind of natural polymer organic matter with poor biochemical degradation and complex composition, which is widely found in papermaking wastewater. Shao *et al.* developed a flexible electrode with Ti/PbO_2 shell as the main electrode and a large number of Fe_3O_4/Sb-SnO_2 particles as the auxiliary electrode to treat papermaking wastewater, which has excellent degradation effect on lignin. After 4 h of reaction, the COD removal rate reached 74%–82.84% [76]. Chai *et al.* used Mp-SnO_2/BDD thin film electrode to degrade wastewater containing chlorogenic acid (CA). After 240 min of treatment, the removal rates of CA and COD by Mp-SnO_2/BDD reached 95% and 90%, respectively, and the degradation rate constant was 1.6 times that of BDD [77].

Benzophenone-3 (BP-3) is one of the most widely used UV absorbers in sunscreens. With the increasing use of sunscreens, it is estimated that 14,000 tons of BP-3 are released into the water environment every year. Zhou used Ti/SnO_2-Sb/Ce-PbO_2 anode to study the electrochemical degradation of BP-3 and its metabolite 4-OH-BP. The initial concentrations of both pollutants were 10 mg/L. At a current density of 25 mA/cm^2 for 30 min, the degradation rates of BP-3 and its metabolite 4-OH-BP were 91.6% and 96.5%, respectively. The addition of Cl^- in the solution significantly promoted the electrochemical degradation of BP-3 and 4-OH-BP. The promotion effect of Cl^- may be due to the direct oxidation of the anode surface or the reaction with $^{\bullet}OH$ to generate active chlorine species (such as Cl_2, ClO^-, ClO_2^- and HClO) [32]. Ruthenium dioxide (RuO_2) is an important electrocatalytic material for electrochemical oxidation of organic compounds due to its high chemical stability, strong electrocatalytic activity and excellent electrical conductivity (1 S/cm). The removal rate of methyl orange and ceftriaxone by RuO_2/TiO_2/nano-graphite anode reached 87.5% and 97.3% at 30 min and 100 min, respectively [78]. Among antibiotics, chloramphenicol (CAP) plays an important role in the prevention and treatment of animal diseases. Its derivatives thiamphenicol (TAP) and florfenicol (FF) have been widely used in animal husbandry. However, TAP and FF have blood toxicity, embryo toxicity and strong immunosuppressive effects, and even affect the physiological functions of animals, plants and microorganisms. The electrocatalytic reduction of thiamphenicol (TAP) and florfenicol (FF) was studied by multi-walled carbon nanotubes modified electrode. After 24 h of electrocatalytic reduction at –1.2 V bias, the removal rates of TAP and FF at 2 mg/L were 87% and 89%, respectively. The removal rate constants of TAP and FF were 0.0837 h^{-1} and 0.0915 h^{-1}, respectively [79].

4.2. Inorganic pollutants

Inorganic pollutants can be divided into two categories: metal inorganic pollutants and non-metal inorganic pollutants. Metal inorganic pollutants mainly include heavy metals such as lead, mercury, cadmium, chromium, zinc, copper, *etc*. Which are usually characterized by strong toxicity and long residual cycle. Nonmetallic inorganic pollutants include acids, alkalis, ammonia, chlorine and other compounds. These inorganic pollutants also have potential threats to the environment and human health. Effective governance measures are needed to strengthen monitoring, control, prevention and treatment in order to protect the environment and human health.

4.2.1. Metal inorganic pollutant

With the rapid development of mining, chemical industry, electroplating, metal products, electronic equipment, leather and other industries, the proportion of heavy metal ions such as chromium, lead, zinc, nickel and copper discharged into water is increasing. Unlike organic pollutants, heavy metal industrial wastewater is non-biodegradable, and heavy metal ions usually accumulate in organisms, posing a serious threat to the health of humans and other organisms. Table S2 (Supporting information) summarizes the removal effect of different inorganic pollutants based on flexible electrodes. Yao *et al.*, by simultaneously indirect electroreduction of Cr(VI) and *in-situ* precipitation of Cr(III), at a current density of 10 mA/cm^2, the maximum removal rate and total removal rate of Cr(VI) within 12 h reached 80.5% and 79.4%, respectively [80]. Shi *et al.* applied PB@CNF electrode to the highly selective recovery of K^+ in simulated seawater. In the presence of high concentrations of K^+, Na^+, Ca^{2+} and Mg^{2+}, the recovery rate of K^+ was as high as 73.4% [81]. Liu simultaneously removed a variety of heavy metal ions coexisting in water through a three-dimensional graphene oxide electrode (CF-GO). At a concentration of 1000 ppb, the removal rates of copper, cadmium and lead were 97.7%, 97.3% and 98.5%, respectively. When the concentration was 10,000 ppb, the removal rates at 20 V were 96.5%, 94.8% and 98.5%, respectively [82]. Ali used iron sulfide modified graphite felt coated reduced graphene oxide (FeS@rGO) nanocomposites as a cathode to achieve simultaneous reduction and power generation of Cr(VI). When the concentration was 15 mg/L, the removal rate of Cr(VI) was 100%, and the maximum power density of MFC-FeS@rGO was 154 mW/m^2, which was 328% higher than that of MFC-blank (36 mW/m^2) [83].

4.2.2. Nonmetallic inorganic pollutant

Nitrate (NO_3^-) pollution in groundwater and surface water resources not only destroys ecosystems, but also leads to eutrophication and serious human health problems. Nitrogen in water usually exists in the form of nitrate (NO_3^-), nitrite (NO_2^-) or ammonium (NH_4^+). Nitrate pollution in surface water and groundwater is mainly caused by human activities, including fertilizer runoff from farmland, rainwater runoff from urban surface, and discharge of sewage or treated wastewater [84]. In recent years, electrochemical nitrogen reduction reaction (NRR) has attracted wide attention because it can convert nitrogen into ammonia under environmental conditions. From the perspective of energy and environment, it is very ideal to convert NO_3^- to NH_3 by electrochemistry, and to achieve nitrate wastewater treatment and ammonia production at the same time [85,86]. However, the cleavage of N≡N bond in N_2 requires 941 kJ/mol, and the decomposition of NO_3^- into deoxygenated species requires only 204 kJ/mol [87]. Electrochemical NRR technology faces the dilemma of low ammonia yield (rNH_3) and faradaic efficiency (FE). In alkaline medium, the following reactions mainly occur (Eq. 6):

$$NO_3^- + 6H_2O + 8e^- \rightarrow NH_3 + 9OH^- \tag{6}$$

Copper is considered to be the best promoter for nitrate electroreduction due to its low cost and relatively high activity. The Cu@Cu_{2+1}ONWs/CP was used as the working electrode, and the electrolysis was carried out at the optimal applied potential of −1.2 V (*vs*. SCE). After 120 min of reaction, the conversion of NO_3-N was 78.57% and the selectivity of NO_3-N was 76% [88]. The nitrate in water was removed by electrochemical process with Cu/GO/Ti as cathode. After electrolysis at a current density of 15 mA/cm^2 for 4 h, the NO_3-N conversion rate can reach 91.5% [89]. Wang *et al.* used 3D Cu nanobelts as cathodes to remove nitrate from water by electrochemical process. They added 0.07 mol/L NaCl to the solution to introduce Cl^-, and electrolysis was performed at an applied voltage of −1.4 V (*vs.* Ag/AgCl). After 30 min, the removal rate of NO_3-N reached 91.5%, and the removal rate of TN reached 99.6% after 120 min [90]. The NO_3^- degradation rate of Cu_3P/CF-400 composite electrode reached 97.7% under the initial condition of 50 mg N L^{-1} NO_3^- after electrolysis for 5 h at the cathode potential of −1.2 V (*vs.* Ag/AgCl). After 8 cycles of experiments, Cu_3P/CF still maintained its electrocatalytic activity [91].

4.3. Organic -heavy metal compound pollution in wastewater

With the increasing complexity of wastewater composition, the treatment of multi-component wastewater has been widely studied. Among them, the removal of coexisting heavy metals and organic matter has received more attention. If not handled properly, the resulting pollutant-rich water may flow into surface water or seep into groundwater, posing a serious threat to human life and drinking water sources. Table S3 (Supporting information) summarizes the removal effect of organic-heavy metal compound pollution in wastewater based on flexible electrodes. Zhang *et al.* used polypyrrole/reduced graphene oxide aerogels (PGAs) as particle electrodes and applied them to a three-dimensional electrode system for simultaneous reduction of hexavalent chromium (Cr(VI)) and bisphenol A (BPA). The results showed that when the solution pH was 3, the concentrations of hexavalent chromium and bisphenol A were 80 mg/L and 20 mg/L, respectively. The removal rates of Cr(VI) and BPA within 30 min were 98.52% and 98.00%, respectively [92]. Under acidic conditions, $Cr_2O_7{}^{2-}$ has a high reduction efficiency for Cr(VI), so H^+ is beneficial to the reduction of Cr(VI) (Eqs. 7 and 8) [93]. Hydroxyl radical is one of the most powerful oxidants in BPA degradation, which is related to the pH of the system. Under acidic and alkaline conditions, the system can produce H_2O_2 as oxidant (Eqs. 9-13). At the same time, under acidic conditions, H_2O_2 can also indirectly generate hydroxyl radicals through catalysts and electrode sheets, so BPA degrades faster under acidic conditions than under alkaline conditions [94].

$$Cr_2O_7{}^{2-} + 14H^+ + 6e^- \rightarrow 2Cr(III) + 7H_2O \tag{7}$$

$$^{\cdot}OH + BPA \rightarrow \cdots \rightarrow CO_2 \uparrow + H_2O \tag{8}$$

Acidic conditions:

$$^{\cdot}O_2 + 2H^+ + 2e^- \rightarrow H_2O_2 \tag{9}$$

$$M_r + H_2O_2 + H^+ \rightarrow {}^{\bullet}OH + H_2O \tag{10}$$

Alkaline conditions:

$$O_2 + H_2O + 2e^- \rightarrow HO^{2-} + OH^- \tag{11}$$

$$HO^{2-} + H_2O \rightarrow H_2O_2 + OH^- \tag{12}$$

$$M_r + H_2O_2 \rightarrow M_oOH + OH^- \tag{13}$$

(M_r is reduced metal catalyst, M_o is oxidized metal catalyst)

Humic acid (HAC) is the most common natural organic matter, which is widely found in soil and aquatic environment. Hexavalent chromium (Cr(VI)) is considered to be the second largest heavy metal in the environment. These two substances usually coexist in the environment. Yin *et al.* prepared PdNPs/AC/BDD thin film electrode. The initial concentrations of HAC and Cr(VI) were 100 mg/L and 20 mg/L, respectively. After electrolysis for 8 h, the degradation rates of HAC and Cr(VI) reached 86.6% and 90%, respectively. Even after five repeated treatments, the removal efficiency of TOC and total Cr was as high as 80% [95].

Veterinary antibiotics (such as tetracycline) and some trace heavy metals (such as arsenic, copper and chromium) are widely used as feed additives in many livestock and poultry farms around the world. The combined pollution system of antibiotics and heavy metals is easy to produce synergistic effect and transform into more toxic antibiotic-heavy metal complexes [96,97]. Bai coupled the electro-catalytic anode (ECA) process with the α-MnO_2 activated PMS process to form an ECA/α-MnO_2/PMS system for the removal of composite pollutants (TC-Cr(VI)) in wastewater. Organic matter is oxidized at the anode and heavy metals are reduced at the cathode. When the concentrations of TC and Cr(VI) were 20 mg/L and 5 mg/L, the removal rates of TC and Cr(VI) were 90.12% and 92% after 5 h, respectively. The main existing forms of Cr(VI) in the solution are $Cr_2O_7{}^{2-}$, $HCrO_4{}^-$, $CrO_4{}^-$ and $HCr_2O_7{}^-$. Under low pH conditions, Cr(VI) mainly exists in the form of $HCrO_4{}^-$ and $Cr_2O_7{}^-$ (Eqs. 14 and 15) [98]:

$$HCrO_4^- + 7H^+ + 3e^- \rightarrow Cr^{3+} + 4H_2O \tag{14}$$

$$Cr_2O_7^- + 14H^+ + 6e^- \rightarrow 2Cr^{3+} + 7H_2O \tag{15}$$

Based on the designed electrochemical double-chamber system, Long *et al.* used three-dimensional reduced graphene as the electrode material to establish a phased and efficient combined antibiotic and heavy ion pollution advanced treatment process. In the anode chamber, TC–Cu combined pollution was degraded into TC and Cu ions, TC was oxidized into small molecular organic matter, carbon dioxide and water, and Cu ions were reduced in the cathode chamber. The removal rates of tetracycline in 20 mg/L and 50 mg/L composite pollutants were 92% and 82%, respectively, and the removal rates of Cu^{2+} were 99% and 100%, respectively. The removal rates of Cu^{2+} and TOC were 89% and 63%, respectively [99].

Shi *et al.* grows on the carbon felt to form a bio-flexible electrode to construct a bio-cathode microbial electrolysis cell (BCMECs). When the applied voltage was 0.9 V and the initial concentration was 20 mg/L, the removal rates of Cu, Pb, Zn, Cd and COD were 98.76%, 98.01%, 73.58%, 84.39% and 77.55%, respectively [100]. Zhao prepared almond shell biochar-based materials with highly dispersed graphene nanosheet structure by using hydrochloric acid and potassium ferrate. Because Fe-AASB has graphene nanosheet structure, abundant oxygen-containing functional groups and mixed iron species, the electrocatalytic reaction at the particle-electrode interface significantly aggravates the electrochemical oxidation reaction, which is beneficial to the effective removal of heavy metals and organic pollutants. After 180 min of reaction, the removal rates of Cu(II), COD and TOC reached 96.8%, 92.5% and 86.2%, respectively [101]. Thamauluslvan *et al.* prepared a robust porous laser-induced graphene (LIG) electrode and demonstrated the electrochemical removal of iodohexanol and heavy metal ion chromium(VI). The optimized LIG electrode showed higher hydrogen peroxide generation. Under the condition of applied voltage of 3 V, the removal rate of Cr(VI) reached 90% after 6 h, and the removal rate reached 95% at pH 2 after 8 h [102].

5. Summary and future prospect

5.1. Summary

This review provides an overview of the development, preparation methods, and application of flexible electrodes in the removal of organic-inorganic pollutants from water. It also addresses the research needs of flexible electrodes in future environmental governance. Several key considerations must be emphasized during the construction of flexible electrodes:

(1) Material selection: Flexible electrodes typically consist of materials with flexible properties and inorganic materials with catalytic properties. Therefore, it is crucial to choose materials that possess elastic deformation ability, excellent electrical conductivity, and catalytic properties.

(2) Process flow optimization: The production process of flexible electrodes is complex, involving multiple steps such as material preparation, dispersion of conductive phases, and the formation of cross-linked networks. It is important to comprehensively evaluate these steps and optimize the overall process flow.

(3) Design of electrode structure: The microstructure of flexible electrodes can be optimized according to the composition and arrangement of electrode materials, such as constructing nanoarray structure, core-shell structure, porous structure and layered structure. These unique structures can provide multiple current channels, increase the contact area between the electrode and the electrolyte, and improve ion and charge transport. At the same time, the advantages of different materials are taken into account to improve the conductivity, flexibility and stability of the electrode.

5.2. Future prospect

Although many studies have demonstrated the feasibility of advanced oxidation technology in wastewater treatment, most experiments are carried out under controlled laboratory conditions, allowing manipulation of water matrix and pollution conditions. In order to evaluate the practical applicability of the advanced oxidation process, it is necessary for researchers to use actual wastewater samples to evaluate its effectiveness. In addition, continuous efforts are needed to discover and develop more powerful flexible electrodes to enhance the performance of wastewater treatment. The design of multifunctional advanced oxidation process reaction device provides a potential solution for the combination of sewage purification and energy production. However, the development of flexible electrodes faces many challenges in research and technology:

(1) Improve the flexibility and adaptability of the electrode: The flexible electrode can be attached to the internal surface or curve part of the sewage treatment system, so as to better contact the target substances and pollutants in the wastewater and achieve higher contact efficiency. The design of flexible electrodes that can be bent, stretched and twisted according to needs can adapt to processing equipment of various shapes and sizes, providing better adaptability and close fit.

(2) Effective loading of active materials: The loading of active materials is a key factor to determine the overall performance and practicability of the electrode. Reasonable selection and loading of appropriate active materials to ensure close contact between active materials and current collectors can extend the functions of flexible electrodes to different fields and applications.

In addition, the combined application with other wastewater treatment technologies, such as adsorbents, membrane separation, *etc*., can also improve the efficiency and comprehensive performance of wastewater treatment. In summary, the future development of flexible electrodes in wastewater treatment has great potential for promoting environmental governance practices.

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.

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Supplementary materials

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