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REVIEW

Application of modified graphite felt as electrode material: a review

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Abstract
Graphite felt is a felt-like porous material made of high-temperature carbonized polymers. It is widely used in electrode materials because of its good temperature resistance, corrosion resistance, large surface area and excellent electrical conductivity. In this paper, the surface functional group modification is of graphite felt electrodes (mainly nitrogen doping modification, nitrogen–sulfur or nitrogen–boron co-doping modification) and surface catalytic modification (metal/ion surface modification and metal oxide surface modification as Main). There are two main methods and research progresses to improve the performance of graphite felt electrodes, and the comprehensive performance of surface functional group-modified graphite felt electrodes and surface catalytically modified graphite felt electrodes are compared respectively. The results show that both surface functional group modification and surface catalytic modification can improve the comprehensive performance of graphite felt electrodes. In this paper, the future development direction of graphite felt activation modification is also prospected.

Keywords Graphite felt · Modification · Electrode material · Flow battery · Electrochemical activity

1 Introduction

With the increase of fossil energy consumption and the aggravation of environmental pollution, people have accelerated the utilization of some renewable new energy (solar energy, tidal energy, etc.) [1]. Due to regional and climatic conditions, renewable energy is intermittent and discontinuous, which limits its wide application [2, 3]. Therefore, it is particularly important to develop a system that is conducive to renewable energy storage [4–6].

Redox flow batteries (RFBS) are an electrochemical energy storage system that separates energy and power [7–9], which has the advantages of safety and environmental protection, long cycle life, and easy energy storage [10, 11], RFBS is an electrochemical energy storage device suitable for electrical energy storage of various scales, and has the potential to become the most promising long-term energy storage technology on the grid scale [12–14]. At present, many researchers at home and abroad are still conducting related research on flow batteries. In the past 40 years of research and development, only the all-vanadium system [15], the iron–chromium system [16] and the zinc–bromine system [17] are close to comprehensive. It is commercialized and has been widely used in the fields of emergency continuous power supply facilities, industrial batteries, electric traction, stand-alone applications, electric load averaging, etc. [18–20].

NASA conducted extensive research on Fe–Cr redox flow batteries in the 1970s, which were the world's first true flow batteries [21]. However, the problem of cross-contamination of iron-chromium flow batteries hinders its further development. Since then, NASA proposed the concept of iron–chromium mixed electrolyte and conducted further research, that is, the positive and negative electrodes are made of ferrous chloride, chromium chloride and hydrochloric acid. Mixed solution, which effectively avoids cross-contamination of active substances in the electrolyte, and the electrolyte after long-term use can be mixed to obtain a new electrolyte. A typical iron–chromium flow battery system consists of a flow circulation system, a membrane separator, two electrodes, and two external reservoirs for the dissolved active

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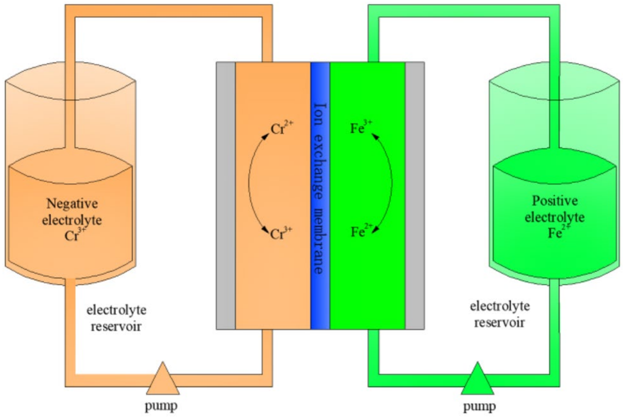


Fig. 1 The role of iron-chromium redox flow battery in energy storage [24]

electrolyte [22, 23]. The role of iron–chromium flow batteries in energy storage is shown in Fig. 1 [24]. During the charging process, the Fe^{2+} of the positive electrode is oxidized to Fe^{3+}, and the Cr^{3} + of the negative electrode is reduced to Cr^{2+}. During the discharge process, the above-mentioned reactions will occur in the opposite process. The electrochemical reaction of the Fe–Cr flow battery during the charge–discharge cycle is shown in Eqs. 1–3 [25]:

$$\text{Positive electrode:}\quad Fe^{2+} - e^{-} \underset{\text{discharge}}{\overset{\text{charge}}{\Leftrightarrow}} Fe^{3+} \quad E^{O} = +0.77\ \text{V vs. SHE} \tag{1}$$

$$\text{Negative electrode:}\quad Cr^{3+} - e^{-} \underset{\text{discharge}}{\overset{\text{charge}}{\Leftrightarrow}} Cr^{2+} \quad E^{O} = +0.41\ \text{V vs. SHE} \tag{2}$$

$$\text{Overall reaction:}\quad Fe^{2+} + Cr^{3+} \underset{\text{discharge}}{\overset{\text{charge}}{\Leftrightarrow}} Fe^{3\,+} + Cr^{3+} \quad E^{O} = +1.18\ \text{V vs. SHE} \tag{3}$$

The key materials of flow batteries include electrodes, membranes, electrolytes, etc. [26–28]. Among them, the commonly used electrode materials are carbon materials, including graphite plate, graphite felt, carbon felt, etc. [29, 30]. Mainly because carbon materials have the advantages of good electrical conductivity and corrosion resistance, they are widely used in flow batteries.

Electrochemical reactions occur on the surface of electrode materials in flow batteries. The hydrophilicity and electrochemical activity of the electrodes will have a direct impact on the electrochemical reactions, which in turn have an important impact on the energy efficiency and power density of the battery. At the same time, the surface of the electrode needs to be continuously washed by the acidic electrolyte, and the electrode needs to have certain mechanical properties, acid resistance and good chemical stability. An ideal flow battery electrode should have the characteristics of high electrochemical activity, high corrosion resistance, good hydrophilicity, and low cost.

Among many carbon electrodes, polyacrylonitrile graphite felt has the advantages of light weight, good flexibility, high carbon content, high temperature resistance, non-volatile at high temperature, corrosion resistance, small thermal conductivity, and high shape retention. The material of choice for flow batteries [31, 32]. At the same time, the polyacrylonitrile-based graphite felt is highly graphitized, poor in hydrophilicity and electrochemical activity, resulting in low battery energy efficiency and power density. Therefore, the development of electrode materials with high activity and excellent hydrophilic properties to improve the energy efficiency and power density of flow batteries has gradually become a current research hotspot.

Most of the ways to speed up the redox reaction of flow batteries, reduce electrochemical polarization, and improve energy efficiency and power density are by changing the performance of graphite felt electrodes [33–35]. The activation methods of graphite felt are generally divided into two methods: increasing surface functional groups and introducing surface catalytic substances [36–38].

The method of increasing surface functional groups is to use oxidation reaction to etch the surface of graphite felt to increase the specific surface area while introducing active functional groups, such as heat treatment [39], acid treatment [40], electrochemical oxidation [41] and chemical vapor deposition [42]. From previous research conclusions, it can be seen that functional groups can enhance the activity of graphite felt [43], while conductivity and specific surface area also affect its performance [44]. Studies have shown that the increase in the specific surface area of the graphite felt makes the electrolyte mass transfer smoother and provides more active sites for the primary reaction [45, 46]. Qiao et al. [47] introduced hydrophilic nitrogen-containing functional groups and oxygen-containing functional groups through the ammonium sulfate hydrothermal method to optimize the hydrophilicity of the graphite felt electrode. The energy efficiency of the flow battery reached 87.34%, which was 3.91% higher than the original energy efficiency. Yang et al. [48] introduced an efficient modulation of graphite felt electrodes through a boron doping method, and the resulting boron-doped graphite felt electrodes could improve the electrochemical performance of flow batteries when an appropriate amount of dopant was provided. Shanahan et al. [49] increased the surface area, oxygen-containing functional groups and electrolyte wettability of the modified graphite felt by rapidly activating the graphite felt in an acidified potassium permanganate solution for 1 h, thereby improving the vanadium redox flow. Battery performance.

The introduction of surface active substances [50] is to add catalysts that can promote the redox reaction of the electrode surface on the surface of the graphite felt, such as Sb, Cu, Bi, PbO_2, ZrO_2, CoO, carbon nanofibers and carbon nanotubes, etc. [51–57]. Loghavi et al. [58] found that the antimony-modified graphite felt electrode has better hydrophilicity than the bare graphite felt electrode, which has a direct impact on the improvement of electrode performance, as well as improved electro-catalysis and gassing inhibition performance. Feng et al. [59] modified the graphite felt electrode with SnO_2, and the reversibility and electrochemical kinetics of the graphite felt electrode were improved. Compared with the blank battery, the SnO_2-modified graphite felt electrode battery has a larger discharge capacity, higher electrolyte utilization and lower polarization. Liu et al. [60] modified a graphite felt electrode with nano-porous carbon, which has abundant porosity and large specific surface area, which promotes the two-electron oxygen reduction reaction and has a higher current response compared with the original graphite felt and lower charge transfer resistance.

Among the various modified graphite felts reported so far, polyacrylonitrile-based graphite felts have become popular in recent years. However, most researchers focus on whether the modification method improves the electrochemical activity. In fact, depending on the nature of the matrix material, these treatments can have beneficial effects.

2 Modification of surface functional groups

Using the oxidation reaction to etch the surface of the graphite felt to increase the specific surface area while introducing active functional groups can enhance the activity of the graphite felt, thereby improving its electrochemical performance. The basic overview of functional group-modified graphite felt electrodes is shown in Table 1.

2.1 Nitrogen doping modification

Studies have shown that the electrochemical performance can be improved by introducing nitrogen-containing functional groups on the surface of graphite felt [61–63]. Anarghya et al. [64] used nitrogen-doped carbon particles prepared from Bermuda grass to modify the surface of graphite felt electrodes. The experimental results showed that the peak values of the cathodic and anodic currents were increased, and the electrochemical properties were also changed. The data obtained from the Nyquist plot and the equivalent circuit show that the MGF has a low charge transfer resistance, the cell has an average Coulombic efficiency of 95%, a peak power density of 60 mW/cm^2, and good stability over multiple cycles. Youn et al. [65] investigated a facile method to prepare nitrogen-functionalized graphite felt (GF) as electrodes for vanadium redox flow batteries. Nitrogen-functionalized graphite felts were produced by ultrasound-assisted self-polymerization and pyrolysis of dopamine (as shown in Fig. 2). After nitrogen functionalization, the electrode surface changed from hydrophobicity to hydrophilicity, the charge transfer resistance decreased, and the electrochemical performance was improved. Excellent electrochemical performance can be obtained by observing the porosity, specific surface area, etc. of the nitrogen-containing functional graphite felt. In the VRFB single-cell test, all NGF samples showed improved energy efficiency. Specifically, at a current density of 150 mA/cm^2, the energy efficiency (75.5%) of the optimized sample NGF05 (containing 0.5 mM dopamine) was higher than that of GF (69.2%). In addition, the electrolyte utilization of GF is only 58.2% while that of NGF05 reaches 68.1%.

Sang et al. [66] proposed another facile method to prepare nitrogen-doped graphite felt electrodes with high electro-catalytic activity for vanadium–oxygen batteries. The surface of the graphite felt was coated with 1-ethyl-3-methylimidazole dicyanamide, and then heat-treated under N_2 atmosphere to prepare nitrogen-doped graphite felt. 1-Ethyl-3-methylimidazole dicyanamide is an ionic liquid with high nitrogen content, which can be used as an effective precursor for nitrogen doping on graphite surfaces. In the charge–discharge test, the nitrogen-doped graphite felt single cell showed excellent performance. Under the condition of current density of 150 mA/cm^2, the surface of graphite felt coated with 20% electrolyte solution (GF-ED20) increased its discharge capacity by 3 times, which was comparable to that of graphite felt with oxygen-containing functional groups (GF-REF) prepared by conventional heat treatment, the energy efficiency is improved by 10%. The improved performance is due to the high nitrogen content of the graphite felt, which also enhances the electro-catalytic activity for the vanadium redox reaction. Dinesh et al. [67] coated N-doped carbon spheres (NDCS) on the surface of graphite felt electrodes, which can significantly improve the performance of iron-based flow batteries. NDCS was synthesized by one-step hydrothermal method using glucose and ammonia water as precursors, and NDCS was prepared on the surface of graphite felt electrode by electrostatic spraying

Table 1 Basic overview of functional group-modified graphite felt electrodes

Electrode modification method	Detection conditions	Properties of unmodified graphite felt electrode	Properties of modified graphite felt electrodes
Nitrogen-doped carbon particle modified graphite felt electrode [64]	Current density 20 mA/cm^2	Coulombic efficiency 37.82%, energy efficiency 12.61%	Coulombic efficiency 92.29%, energy efficiency 49.19%
Dopamine pyrolysis modified nitrogen-functionalized graphite felt [65]	Current density 150 mA/cm^2	Energy efficiency 69.2%, electrolyte utilization 58.2%	Energy efficiency 75.5%, electrolyte utilization 68.1%
Nitrogen doping modified graphite felt electrode [66]	Current density 150 mA/cm^2	About voltage efficiency 69%, energy efficiency 68%	About voltage efficiency 80%, energy efficiency 78%
Nitrogen-doped carbon spheres modified graphite felt electrode [67]	Current density 20 mA/cm^2	About coulombic efficiency 62%, voltage efficiency 65%, energy efficiency 40%	About coulombic efficiency 90%, voltage efficiency 82%, energy efficiency 73%
Nitrogen-doped reduced graphite oxide modified graphite felt electrode [68]	Current density 80 mA/cm^2	Coulombic efficiency 92.62%, voltage efficiency 81.43%, energy efficiency 75.42%	Coulombic efficiency 92.09%, voltage efficiency 82.62%, energy efficiency 76.08%
Sulfur-nitrogen co-doped graphene modified graphite felt electrode [70]	Current density 80 mA/cm^2	About energy efficiency 85.37%	Energy efficiency 85.37%
S/N co-doped GQD graphite felt modified electrode [71]	under the same conditions	Discharge capacity 141 mAh	Discharge capacity 155 mAh
Nitrogen-doped porous carbon modified graphite felt electrode [69]	Current density 12.5 mA/cm^2	H_2O_2 Generation rate 0.067 mg h^{-1} cm^{-2}	H_2O_2 Generation rate 0.74 mg h^{-1} cm^{-2}
Doped nitrogen/boron modified graphite felt electrode [72]	Under the same conditions	H_2O_2 generation rate 9.39 mg/l, current efficiency 1.61%	H_2O_2 generation rate 152.8 mg/l, current efficiency 70.3%

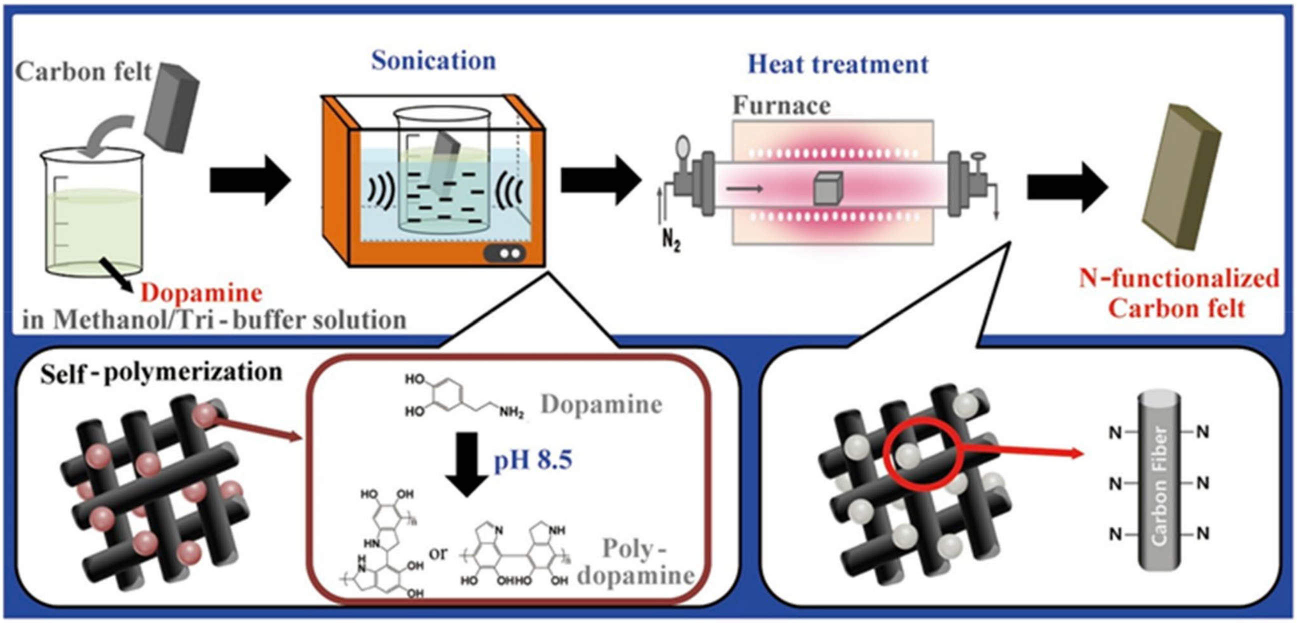


Fig. 2 Preparation of NGF electrode via ultra-sonication-assisted self-polymerization and subsequent pyrolysis [65]

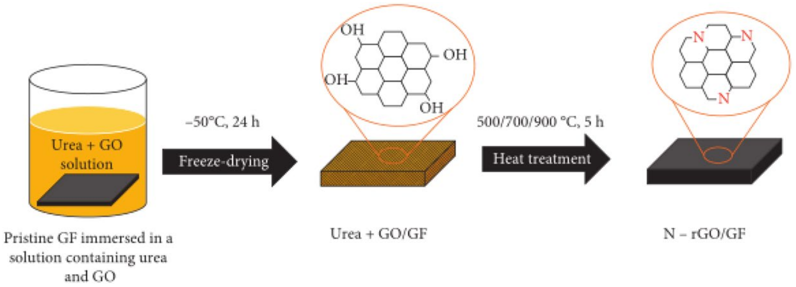
method. During 15 cycles at a current density of 30 mA/cm^2, the cell achieved an average coulombic efficiency of 93%, a current efficiency of 72%, and an energy efficiency of 68%. Since the surface of the graphite felt is nitrided, its electrochemical activity is also improved. To improve the positive reaction kinetics, Li et al. [68] used a combination of freeze-drying and pyrolysis (as shown in Fig. 3) to prepare a nitrogen-doped reduced graphite oxide modified graphite felt (N-rGO/GF) electrode. The combined method of freeze-drying and pyrolysis can make N-rGO uniformly dispersed on the surface of glass fiber electrode (as shown in Fig. 4), with good catalytic activity. The results show that the composite electrode has good electrochemical performance at a pyrolysis temperature of 900 °C, and the catalytic activity and electrochemical reversibility of VO^{2+}/VO_2^+ positive ions are significantly improved, indicating that the composite electrode has potential in improving VRFB performance. Application value.

Fy et al. [69] investigated a novel modified graphite felt (GF) as an electro-Fenton (EF) cathode material, which supported nitrogen-doped porous carbon (NPC) with a zeolite-type imidazolium salt framework-8 (ZIF-8) is a carbon precursor. The cathode modified with phenol (50 mg/L) by NPC can greatly remove organic carbon in wastewater. When the optimized conditions are 120 min, the mineralization rate reaches 100%, and the total organic carbon reaches 82.61%. After 5 cycles, the NPC cathode remained stable. The experimental results show that the electrochemical activity of the modified graphite felt is improved, which is due to the uniform distribution of nanoparticles on the fiber surface and the promoting effect of sp2 carbon and graphitic N between the carbon surface and oxygen molecules. Optimization of fabrication parameters, such as current, pH, and NPC loading, improved the efficiency of a novel cathode for in-situ electrochemical production of hydrogen peroxide, providing a potential material for degrading organic pollutants.

2.2 Nitrogen-sulfur, nitrogen–boron co-doping modification

In previous reports, it was found that the synergistic effect of heteroatoms in nitrogen–sulfur or nitrogen–boron co-doped structures increased the electrochemical activity of graphite felt. Li et al. [70] used thiourea as the sulfur and nitrogen source, and modified double nitrogen-doped graphene on graphite felt electrodes by hydrothermal method for vanadium redox flow battery. The results show that the introduction of nitrogen and sulfur on the surface of graphite felt can introduce functional groups and increase the surface area for its reaction. At a current density of 80 mA/cm^2 (as shown in Fig. 5), the energy efficiency, power density, discharge capacity and retention capacity were significantly improved. These results indicate that nitrogen–sulfur co-doped graphene well improves the activity of graphite felt electrodes,

Fig. 3 Schematic diagram of the N-rGO/GF fabrication process [68]



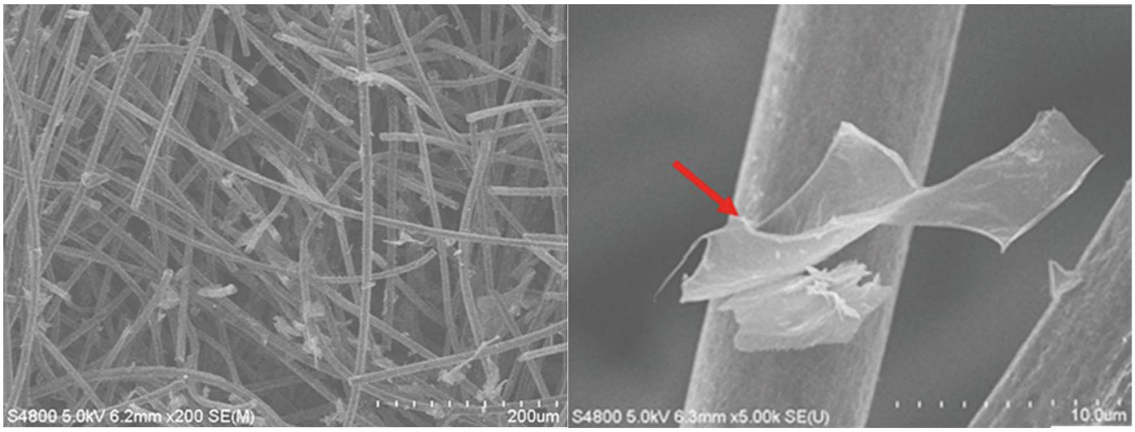


Fig. 4 Surface morphology of N-rGO-modified GF electrode (**a**) and detailed view of the N-rGO absorbed on GF electrode. Te red arrow in (**b**) points to the absorbed N-rGO [68]

providing an effective way to enhance the performance of vanadium redox flow batteries.

Mcd et al. [71] used sulfur/nitrogen co-doped graphene quantum dots (GQDS) to modify graphite felt electrodes to improve the electrochemical performance of vanadium redox flow batteries. The average diameter of S/N co-doped GQDs is 6.2 nm, and various performances of vanadium redox flow batteries are improved to varying degrees under the action of S/N co-doped GQDs. This is mainly due to the introduction of GQDs doped with O functional groups, lattice N atoms and S. Under their combined action, the charge transfer in the electrolyte is accelerated, and the catalytic activity of the vanadium redox surface is enhanced. Thus, the multi-level S/N co-doped GQD/GF electrodes paves the way for engineered electrode nanostructures that enhance the catalytic activity and durability of redox flow batteries.

To promote the generation of dimethyl phthalate (DMP), Ding et al. [72] developed a modified graphite felt cathode doped with nitrogen and boron and applied it in a hydrogen peroxide-coagulation system. After simple modification, the cathode H_2O_2 yield was increased from 9.39 to 152.8 mg/l, and the current efficiency was increased from 1.61 to 70.3%. Under the optimal conditions of pH 5, cathode potential of -0.69 V, and electrode spacing of 1 cm, DMP was completely degraded within 2 h, and the TOC removal rate reached 80%. Through quenching experiments, the possible mechanism of the synergistic effect of electro-reagents and electro-coagulation in the peroxide-coagulation system was revealed (as shown in Fig. 6). The application prospect of the system in landfill leachate and domestic sewage treatment was studied. The removal rate of the system reached 50% and 61% respectively within 2 h. The system adopts a simple modified cathode and has a good application prospect.

3 Modification of surface catalytic materials

Modification of graphite felt by metal or metal oxide can improve its electrochemical performance. The basic overview of surface catalytic material modification is shown in Table 2

Vanadium redox flow batteries stand out among various electrochemical energy storage technologies due to their good operational flexibility and scalability. However, traditional electrodes with low electrochemical activity, such as carbon felt and graphite felt, hinder the interfacial charge transfer process, resulting in considerable over-potential loss, which greatly reduces the energy efficiency and voltage efficiency of the electrode.

Xia et al. [73] used a simple electrodeposition technique to prepare the composite electrode, and successfully obtained high electrochemical activity. The Pb/CF electrode has good electrochemical activity for the VO_{2+}/VO_2^+ redox couple in the battery. At 100 mA/cm^2, the battery voltage efficiency reaches 90% and the energy efficiency also reaches 88%. In addition, due to the uniform distribution of lead

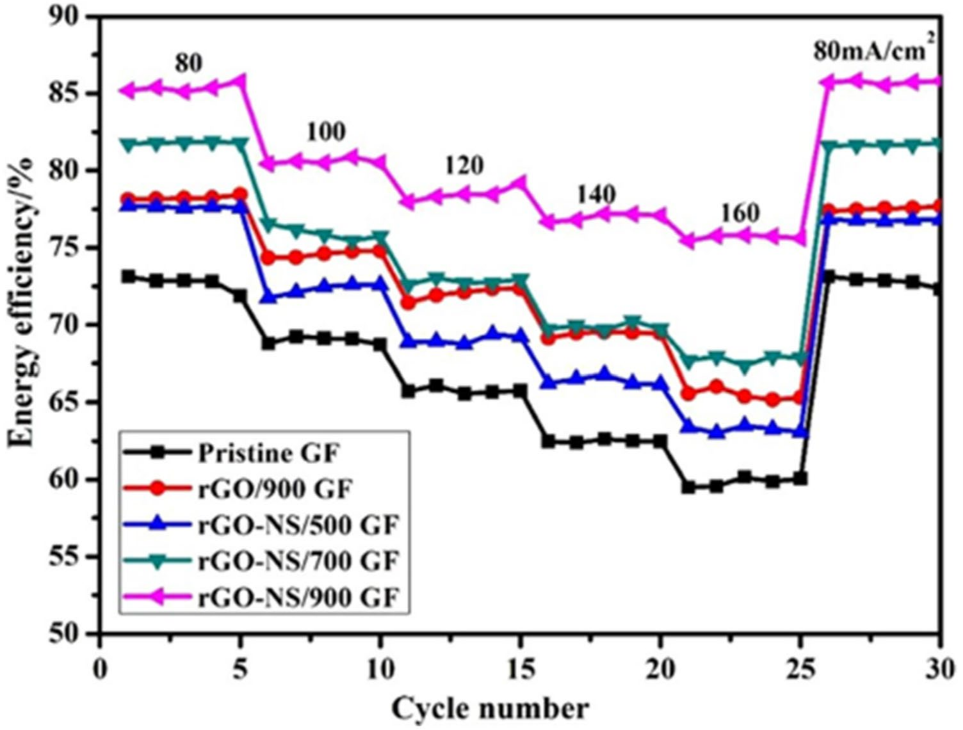


Fig. 5 The energy efficiency for different samples in the current density range of 80–160 mA/cm^2 [70]

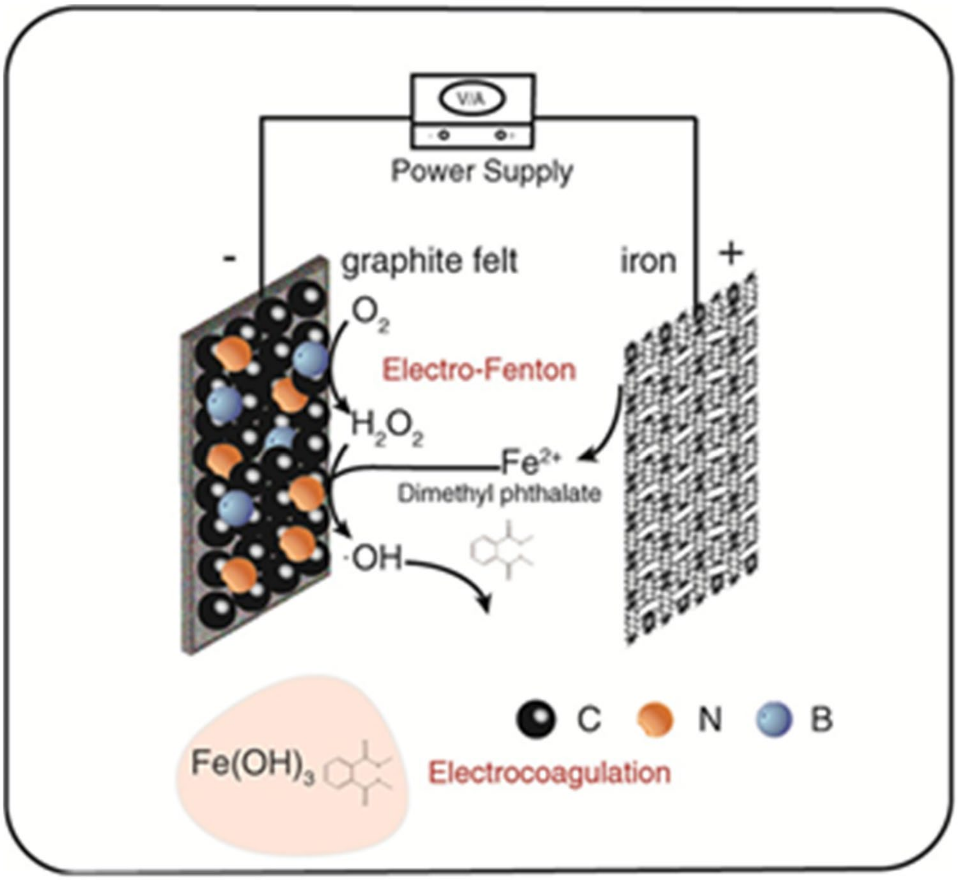


Fig. 6 The proposed mechanisms for DMP degradation in PC system with modified cathode material [72]

particles on the surface of the graphite felt, the long-term stability of the graphite felt battery with the Pb/CF electrode is significantly better than that of the pristine graphite felt battery due to the redox-mediated catalysis. Furthermore, three single cells were combined into a stack to estimate the reliability of the redox-mediated Pb/CF electrode application in a large scale (as shown in Fig. 7). This technique of preparing electrodes from high-performance and facile redox-mediated reactions is expected to provide new ideas for industrial electrode design.

Among various metal halide redox flow batteries, polyhalide-based hybrid redox flow batteries are widely used in the field of long-term fixed charge storage due to their long cycle life and high energy density. Zinc–bromine flow battery systems have attracted much attention due to their reasonable battery voltage, energy density, and lifetime. However, the main reason why the performance of the battery cannot be improved is because of the generation of bromine vapor during the charging process, and the poor kinetics of the bromine oxidation reaction on the electrode is related to this flow system. To improve the reaction kinetics of the zinc–bromine battery, Mariyappan et al. [74] introduced noble metal platinum particles on the graphite felt electrode, and the platinum particles accelerated the electron transfer rate in the electrode. The graphite felt was modified by a simple synthesis process. The pt content was optimized by improving the reaction kinetics of br_2/br^-. Mariyappan et al. [75] also used low-loaded platinum as the electro-catalyst (as shown in Fig. 8), and the br_2/br^- reaction kinetics were better improved due to the deposition of platinum particles. The improvement of the kinetics of the br_2/br^- redox coupling reaction was studied by introducing platinum as the electrode material on the surface of the graphite felt. This method greatly affects the kinetics of the bromine/bromide redox reaction. A Coulombic efficiency of 99.96% is the best average efficiency. Under the condition of 50 mA/cm^2, the energy efficiency of the zinc–bromine-based flow battery was measured to be 88%. Therefore, graphite felt electrodes modified with low loadings of platinum are used as materials to improve br_2/br^- kinetics in most studies.

To improve the catalytic activity of graphite felt (GF) electrodes, Wang et al. [76] proposed to modify the surface of GF with metal oxides. In supported oxides, the presence of nanoparticles significantly enhances mass transfer and reactivity. X-ray photoelectron spectroscopy and contact angle measurements showed that the content and hydrophilicity of oxygen-containing groups increased significantly.

Table 2 Basic overview of surface catalytic material modification

Electrode modification method	Detection conditions	Properties of unmodified graphite felt electrode	Properties of modified graphite felt electrodes
Pb modified graphite felt electrode [73]	Current density 100 mA/cm^2	Voltage efficiency 83.45%, energy efficiency 80.58%	Voltage efficiency 90%, energy efficiency 88%
Platinum modified graphite felt electrode [74]	Current density 40 mA/cm^2	Coulombic efficiency 92.76%, voltage efficiency 70.97%, energy efficiency 65.83%	Coulombic efficiency 94.72%, voltage efficiency 85.35%, energy efficiency 80.42%
Platinum modified graphite felt electrode [75]	Current density 50 mA/cm^2	Coulombic efficiency 89.79%, energy efficiency 67.93%。	Coulombic efficiency 99.96%, energy efficiency 88%
$LaSrO_x$ modified graphite felt electrode [76]	Current density 50 mA/cm^2	Energy efficiency 54.76%	Energy efficiency 61.37%
Cerium modified graphite felt electrode [77]	Current density 30 mA/cm^2	Energy efficiency 27.9%	Energy efficiency 58.4%
Co_3O_4 modified graphite felt electrode [78]	Current density 100 mA/cm^2	Voltage efficiency 83.66%, energy efficiency 80.38%	Voltage efficiency 91.53%, energy efficiency 87.6%
Cobalt oxide modified graphite felt electrode [79]	Current density 150 mA/cm^2	Coulombic efficiency 84.7%, voltage efficiency 66.9%, energy efficiency 56.7%, discharge capacity 185.4 mAh	Coulombic efficiency 89.5%, voltage efficiency 77.6%, energy efficiency 69.4%, discharge capacity 373.9 mAh
WO_3 modified graphite felt electrode [80]	Current density 40 mA/cm^2	Coulombic efficiency 18%	Power density 53 mW/cm^2, coulombic efficiency 64%
Vanadium oxide modified graphite felt electrode [82]	Current density 150 mA/cm^2	Voltage efficiency 57.57%	Voltage efficiency 66.96%
α-MnO_2 modified graphite felt electrode [83]	Current density 4 mA/cm^2	Negligible battery capacity	battery capacity 1.26 mAh/cm^2, cycle capacity retention 99.8%
Mn_3O_4 modified graphite felt electrode [84]	Current density 80 mA/cm^2	Voltage efficiency 81.1%, energy efficiency 78.1%	Voltage efficiency 83.8%, energy efficiency 79.5%

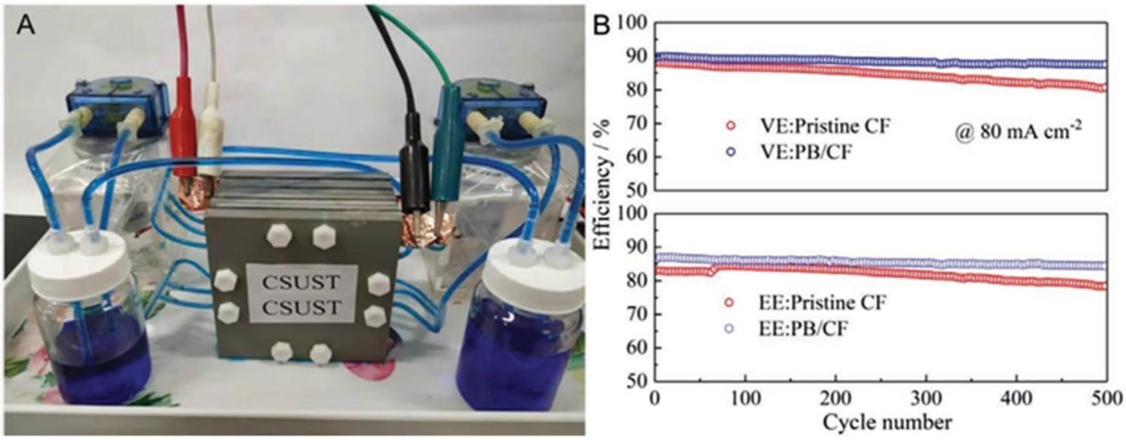


Fig. 7 VRB cell stack. **A** Photograph of a VRB cell stack consisting of three single cells (16 cm.^{2}) and **B** cycling stability of the VRB cell stack with the CF and PB/CF electrodes, respectively [73]

After assembling the electrodes in the battery, the laser/glass fiber electrodes greatly improved the performance of the battery, significantly increasing the capacity and efficiency of the battery. When the current is 50 mA/cm^2, the laser/glass fiber electrode increases the energy efficiency of the battery from 54.76 to 61.37%. In addition, the cyclability test of the system shows that there is no obvious fading phenomenon after 100 cycles, indicating that the $LaSrO_x$/GF electrode has good stability.

The low efficiency of cerium-based redox flow batteries is mainly due to the limitation of the kinetics of the cerium redox reaction. Therefore, it is very important to develop electrode materials that can accelerate the redox reaction. Na et al. [77] proposed an activation method that utilizes

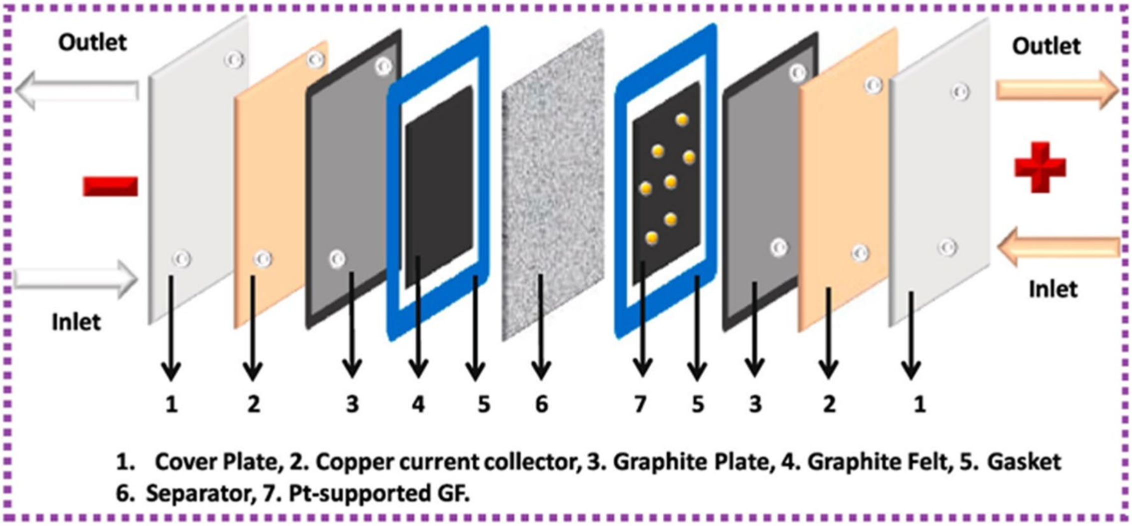


Fig. 8 Exploited view of the Flow cell configuration used in the present investigation [75]

surface functionalization to significantly improve the electrochemical performance of commercial graphite felts. Activated graphite felt as an electrode for cerium redox reaction has good catalytic activity and durability for Ce(IV)/Ce(III) redox coupling. It is found that the combination of oxygen functional groups, especially O–C=O functional groups, on the active graphite felt electrode accelerates the charge transfer rate, thereby improving the performance of the electrode.

Some researchers have found that some inexpensive metal oxides can be used as catalysts for flow battery electrode materials. Mahanta et al. [78] used a low-temperature hydrothermal method to prepare thermally activated graphite felt with Co_3O_4 nanostructures (as shown in Fig. 9). Functional groups on TGF can nucleate Co_3O_4 particles, creating covalent bridges between them. The bridge improves the tunneling of electrons across the electrolyte/electrolyte interface and reduces the over-potential of the vanadium redox reaction. The EEs of the two battery packs after 50 cycles are 84% and 89% for their initial capacity, respectively, and the 3% loss of EE compared to a single cell is mainly due to the additional contact resistance due to cell coupling.

Xiang and Daoud [79] attached cobalt oxide to the surface of graphite felt in the form of a coating. Through electrochemical detection, it was found that the modified graphite felt has good electro-catalytic activity and reversibility, and after 50 cycles of repeated experiments has stability. The coulombic efficiency, voltage efficiency, energy efficiency and discharge capacity of the battery detected by charging and discharging are 89.5%, 77.6%, 69.4% and 373.9mAh, respectively. Compared with the graphite felt battery, the energy efficiency is increased by 12.7% and the discharge capacity is increased by 101.7%. Polarization curve analysis shows that the limiting current density and maximum power density are significantly increased at 95% of the charge state due to the suppression of ohmic polarization. The results show that the modification of light cobalt has a good and stable catalytic effect on improving the electrochemical performance of graphite felt in vanadium redox batteries.

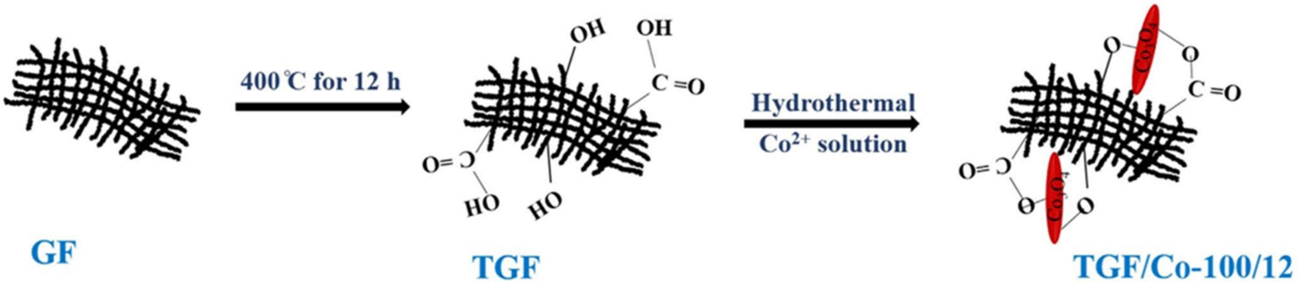


Fig. 9 Schematic illustration of the fabrication of Co_3O_4-modified electrodes [78]

Iron-based flow batteries are of great interest due to their economic viability and environmentally friendly electrolytes. The electrodes and electrolytes used in iron-based redox flow batteries (IRFBs) play a crucial role in the performance of electrochemical energy storage devices. Therefore, it is very necessary to design suitable electrodes and optimize the electrolyte composition. Graphite is one of the suitable electrodes in flow batteries, but it must be modified to improve its electrical properties. Dinesh et al. [80] used WO_3 nanoparticles (WON) for the first time to modify graphite felt electrodes for IRFBs applications. The electrochemical activity of the modified graphite felt electrode was increased by electrochemical detection. Faggiano et al. [81] studied the heat treatment time of copper-based redox flow battery graphite felt, and the polyacrylonitrile-based carbon felt showed excellent stability and the best electrochemical performance after heat treatment at 400 °C for 6 h. Wang et al. [82] prepared a simple and environmentally friendly graphite felt active electrode by introducing vanadium oxide on the surface of the graphite felt. On the activated graphite felt surface, the surface porosity and roughness increased when the calcination temperature and surface area reached 350 °C to 400 °C and 17.11 m^2/g, respectively. At a charge transfer resistance of 0.27 Ω, the polarization of the activated graphite felt decreases with decreasing charge transfer resistance.

Yan et al. [83] designed a simple one-step activated graphite felt containing nitrate to improve the energy storage performance and cycling durability of batteries by increasing the number of oxygen-containing functional groups and mesoporous. Under the condition of 4 mA/cm^2, its area capacity is 1.26 mAh/cm^2, and the cycle capacity retention rate is 99.8%. Furthermore, at a power density of 3.7 mW/cm^2, the cell provides a remarkable energy density of 1.82 mWh/cm^2. This modification strategy and material have great potential for wear-resistant, flexible devices.

For the application of vanadium redox flow battery (VRFB), Chen [84] proposed a simple and effective method for roughening graphite felt (GF) using manganese oxide as an etchant (as shown in Fig. 10). At different current densities, VRFBs with R-GF electrodes exhibit strongly enhanced electrochemical performance in terms of energy efficiency and electrolyte utilization. The large specific surface area of R-GF with abundant oxygen-containing functional groups is extremely favorable for improving VRFB kinetics.

Qi et al. [85] used a one-step hydrothermal method to in-situ grow transition metals Cu and Fe on the surface of a graphite felt electrode without a polymer binder. The electrode exhibits low metal ion leaching rate and good stability. Jiang et al. [86] used metal–organic frameworks (MOFs) to prepare metal oxides and porous carbon nanocomposites for the first time, and used the in-situ growth method for ultra-uniform surface modification of graphite felts. Using a hydrothermal synthesis method, the graphite felt was modified with UiO-66 (Zr-MOF) nanoparticles, which were then converted into porous nanocomposites (ZrO_2@C) by high-temperature carbonization (as shown in Fig. 11). In the 500-cycle test, the flow battery modified with ZrO_2@C/GF has better stability and higher electrolyte utilization than the primary battery. The performance of the studied cells is superior to that reported in previous metal oxide-related studies.

In terms of current efficiency and mass transfer, the current of flow electrodes generally exceeds that of conventional parallel plate electrodes. The high cost and complex fabrication of high-performance electrode materials hinder their widespread application. Chen et al. [87] used graphite felt as the electrode of a flow battery to study the ability

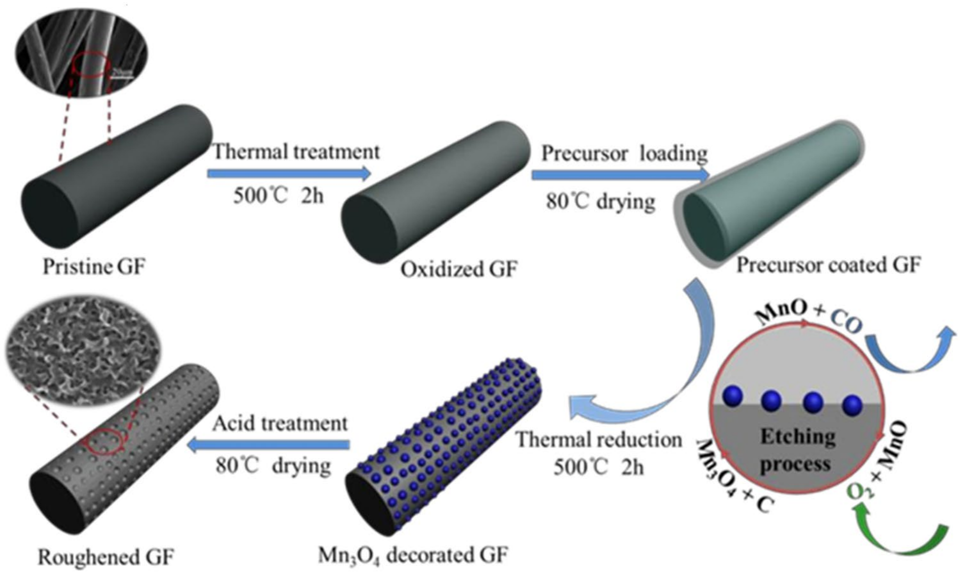


Fig. 10 Schematic of the fabrication process of roughened graphite felt [84]

of the electrode to electro-reduce Cr(VI) in solution. Tests under acidic conditions show that the reduction efficiency of Cr(VI) is very high and can reach 95–100%. The electro-reduction of Cr(VI) is due to the low flow rate and high current in the cell, while the conductivity of Cr(VI) in dilute solution is enhanced by the presence of other metal ions in the solution. Low-valent ions can reduce Cr(VI) better at low flow rate, but these ions reduce the reduction efficiency of Cr(VI) at fast flow rate. In addition, the surface of the graphite felt is hindered from forming an insoluble layer in an acidic environment, which reduces the energy loss of the battery and improves the durability of the graphite felt.

To improve the electro-catalytic activity of pristine graphite felt (GF) for V^{3+}/V^{2+} redox reaction and reduce the influence of hydrogen evolution reaction on battery performance, Xiao et al. [88] used hydrothermal method to load cadmium oxide (CdO) nanoparticles on graphite felt On the surface, modified graphite felt (CdO/GF) was prepared as a high-performance vanadium battery anode. Compared with GF, CdO/GF effectively inhibited the activity of hydrogen evolution reaction. CdO/GF significantly improved the electrochemical activity and reversibility of V^{3+}/V^{2+} redox reaction, and the charge transfer resistance was also significantly reduced, compared with GF, the discharge capacity decay rate of CdO/GF decreased significantly, and the voltage efficiency and energy efficiency increased by about 5% at a current density of 90 mA/cm^2. The catalytic performance of CdO/GF shows good stability during multiple charge–discharge cycles. Li et al. [89] used a combination of impregnation and high-temperature calcination to deposit non-noble metal chromium oxides on the surface of graphite felt to modify the graphite felt electrode (as shown in Fig. 12). In the charge–discharge cycle test, the energy efficiency of the nanopowder assembled with the chromium oxide modified electrode was 87.2%. This provides a possible route for deep eutectic solvent (DES) electrolytes to enhance the properties of nanofibers.

Zhang et al. [90] prepared an active electrode by introducing magnesium oxide (MgO/GF) catalyst into the surface of graphite felt by isometric impregnation method. Mechanistic studies show that MgO/GF has high activity in the ozonation of BPA. Lou et al. [91] developed a three-dimensional porous Ag–Bi electrode to realize the highly selective reduction of CO_2 to formate (as shown in Fig. 13). A galvanic displacement reaction was used to deposit silver particles on the surface of double-layer graphite felt. Compared with the double-coated graphite felt, the reduction selectivity of the silver-double electrode is enhanced. Formic acid is converted from CO_2 under the action of the bimetallic catalytic system, and its selectivity can reach 88% at − 1.6 V. The highly porous structure and large surface area of the electrode facilitates the mass transfer of the electrode, which enables the electrode surface current density to reach 76 mA/cm^2, and the yield is also improved.

Lou et al. [92] studied the reduction and removal of the organochlorine herbicide alachlor by nano-silver modified nickel-coated graphite felt (Ag/Ni@GF). A new type of graphite felt electrode is prepared by coating the surface of the graphite felt with a layer of metallic nickel and then modifying it with silver ions. Through Raman scattering detection, it can be known that the content of PVP as a material for controlling the deposition of silver nanoparticles will not affect the catalytic activity of silver. And the prepared composite electrode is very stable, so it can be reused many times.

In the heterogeneous electro-Fenton (EF) process, transition metal oxide-based carbon composite electrodes with high stability and high activity have broad application

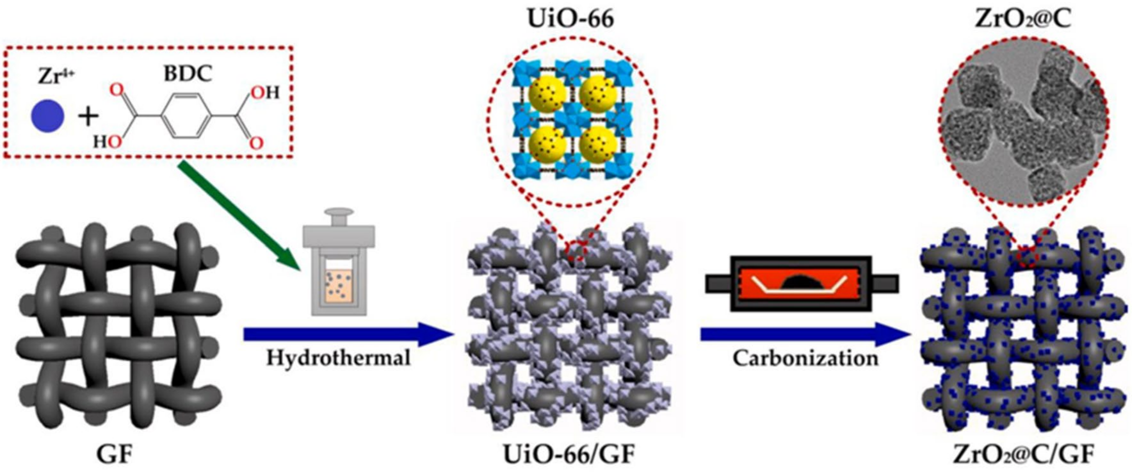


Fig. 11 Schematic representation of the procedure for preparing graphite felt electrode decorated with in-situ grown porous ZrO_2@C nanoparticle derived from metal–organic framework [86]

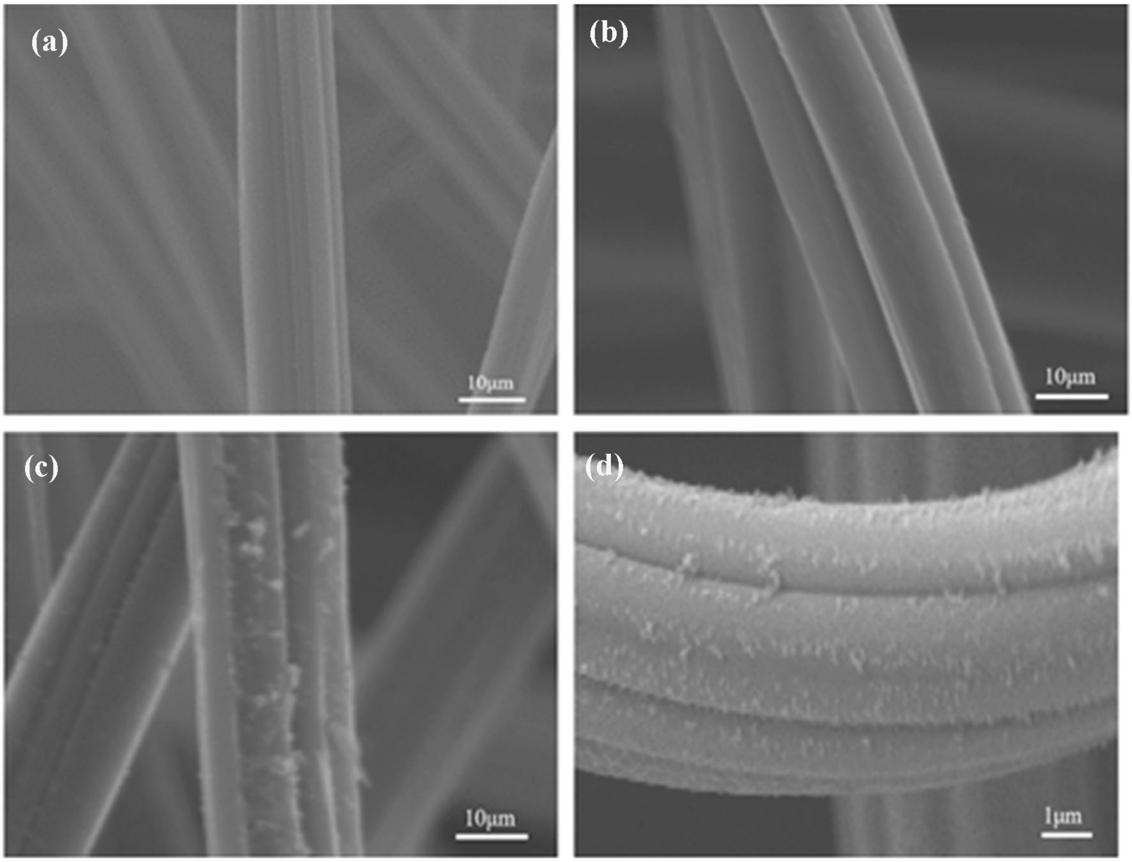
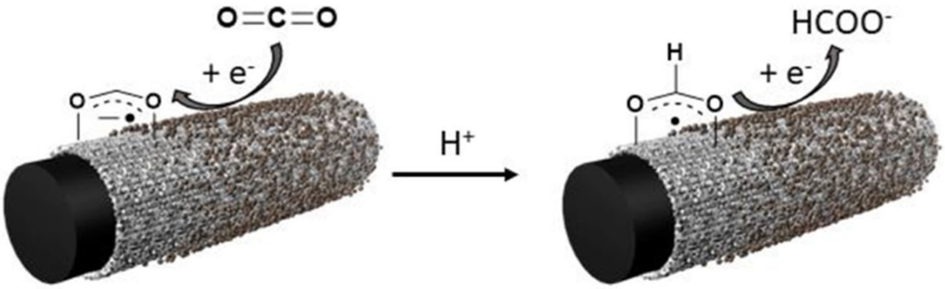


Fig. 12 SEM images of PGF (**a**) and TGF (**b**) and CrTGF (**c**, **d**) [89]

Fig. 13 Proposed mechanism for the selective reduction of CO_2 at the bimetallic electrode [91]



prospects in environmental governance. Cui et al. [93] synthesized Cu/$CuFe_2O_4$(CCFO) with different Cu^0 ratios in one step by solvothermal method. These materials were then used to manufacture modified graphite felts by polytetrafluoroethylene (PTFE) bonding techniques. Importantly, the hydrophobicity of PTFE effectively inhibits metal leaching from the cathode and improves oxygen utilization. During the degradation of EF-based tetracycline (TC), the cathode performance increases with the increase of Cu^0 ratio. Cu^0 enhances the selectivity of the 2-electron oxygen reduction reaction and endows the cathode with abundant electron-rich centers, accelerating the regeneration of active Fe(II), resulting in the rapid conversion of H_2O_2 to hydroxyl radicals. A possible TC mineralization pathway was deduced by liquid chromatography–mass spectrometry analysis, and a catalytic mechanism for the heterogeneous EF process was proposed. The most efficient cathode also showed high stability at pH 3 (0.84 ± 0.11 mg/L Fe and 1.35 ± 0.17 mg/L Cu leaching), with TC removal close to 80% after 5 oxidation cycles, indicating its potential for efficient and durable wastewater treatment in heterogeneous EF technology. Lu et al. [94] designed a three-dimensional bimetallic carbon-based electrode CuNi/multi-walled carbon nanotubes/graphite felt (CuNi/M/GF) for the electrochemical reduction of nitrate by adjusting the electrodeposition potential (As shown in Fig. 14) CuNi/M/GF exhibited excellent corrosion resistance and stability in cycling tests. Yang et al. [95] proposed a batch catalyst electrodeposition method to resolve the conflict between electrodeposition current density and catalyst distribution, and achieved a more uniform and dispersed catalyst distribution inside the porous electrode, thus, compared with the optimized, has higher voltage efficiency and electrolyte utilization. Nariyama et al. [96] introduced redox mediators in the no aqueous electrolyte, and the active materials were charged and discharged without direct contact with the electrodes. A novel high-capacity and high-pressure redox flow battery was fabricated.

4 Conclusion and outlook

Redox flow batteries and to a lesser extent hybrid flow batteries have the advantages of flexible layout (due to separation of power and energy components), long cycle life (because there is no solid–solid phase transition), fast reaction times, and no need for “equilibration” Charging (overcharging of the battery to ensure all cells have the same charge) and no harmful radiation. Some types also offer simple state-of-charge determination (charge-dependent via voltage), low maintenance and tolerance for overcharge/over-discharge. These technical advantages make redox flow batteries ideal for large-scale energy storage.

Electrode materials are one of the key materials that need to be broken through in flow batteries. Graphite felt is widely used as an electrode material for redox flow batteries due to its stable electrochemical performance, high mechanical strength, and large surface area. However, the graphite felt material has poor hydrophilicity and insufficient electrochemical activity, and needs to be modified before being used as a battery electrode.

This paper introduces the surface functional group modification of graphite felt electrode (mainly nitrogen doping modification, nitrogen–sulfur or nitrogen–boron co-doping modification) and surface catalytic modification (metal/ion surface modification and metal oxide surface modification as the main Main) two main methods and research progresses to improve the performance of graphite felt electrodes, mainly focus on improving the specific surface area, hydrophilicity and electrical conductivity of graphite felt, so as to achieve the purpose of speeding up the reaction rate and improving the electrochemical performance of graphite felt electrodes. Judging from the research results obtained so far, the graphite felt electrode still needs to further improve its electrochemical activity to improve the performance of the electrode. At the same time, with the development of the redox flow battery industry, low-cost, high-performance graphite felt modification method suitable for industrial production is still an important development direction of redox flow battery electrode research. In view of this, future research on graphite felt electrodes will mainly focus on the following work:

1. At present, the metals and metal oxides introduced on the surface of the graphite felt are mainly sub-group (B) metal elements. This is an accidental or inevitable conclusion, and no one has given a definite conclusion, so it needs to be systematically studied to clarify;

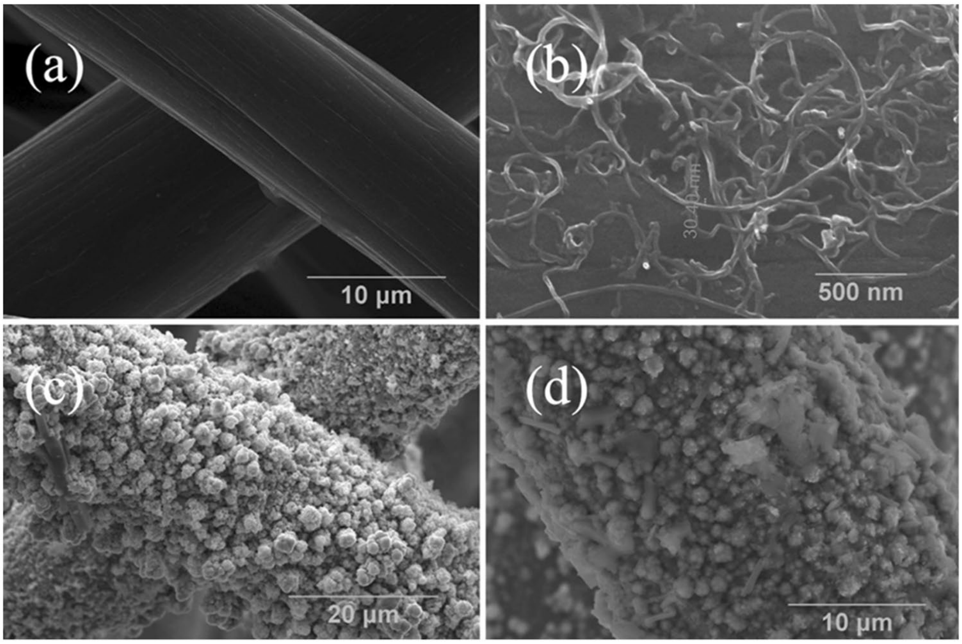


Fig. 14 SEM images of **a** GF, **b** M/GF, **c** CuNi–0.7/M/GF, **d** CuNi–1.3/M/GF [94]

2. In terms of functional group modification, nitrogen doping modification, nitrogen–sulfur or nitrogen–boron co-doping modification is mainly used, and organic functional groups, such as carbonyl group and carboxyl group, are also used for modification, but this modification is due to the introduction of radicals. The lone pair of electrons in the group is caused by other factors, and this kind of modification mechanism has not yet formed a unified theory, so further systematic research is needed;
3. In terms of functional group modification, in addition to single functional group modification, multi-element doping can be introduced (such as –OH doping-SH; –COO-doping-CSS-, etc.). Or whether the co-introduction of multiple functional groups (such as carbonyl + carboxyl) can increase the electrochemical activity of the graphite felt electrode is also worth investigating;
4. The current research on the activation of graphite felt electrodes is limited to experimental research. In the context of digitalization and big data, whether it is possible to establish mathematical models, carry out simulation experiments, and combine theory and experiments to improve graphite felt electrodes. Electrochemical activity;
5. The problem of expanding the application field of subsequent graphite felt modified materials. That is to say, in addition to being used as an active electrode in energy storage systems, the modified graphite felt should also be considered as a cathode material in other applications such as degrading pollutants in water.

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Declarations

Conflict of interest The authors declare that they have no conflict of interest.

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