ISSN: 2710-0588

Research Progress of Bio-electro-Fenton System and Its Application in Wastewater Treatment

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Abstract

The bio-electro-Fenton (BEF) system integrates bioelectrochemistry with Fenton advanced oxidation technology. It can achieve self-sustained electricity generation via anaerobic microbial metabolism and construct an efficient advanced oxidation system relying on in-situ hydrogen peroxide production at the cathode. This system effectively addresses the limitation of conventional biological methods that struggle to treat refractory and highly toxic organic wastewater, thus holding promising application prospects in water pollution remediation. This paper systematically reviews the core reaction mechanisms of BEF systems, elaborates the microbial electrogenic oxidation mechanism at the bioanode as well as the oxygen reduction and Fenton catalytic oxidation mechanisms at the cathode, and summarizes recent research advances in modification and optimization of anode and cathode electrode materials. Emphasis is placed on analyzing the effects of carbon-based modification, nanocomposites and biochar-based electrode materials in boosting electron transfer efficiency and enhancing electricity generation performance and hydrogen peroxide production efficiency. Meanwhile, this paper compares and evaluates the merits and demerits of homogeneous iron-based catalysis and supported heterogeneous catalytic systems. It points out that traditional homogeneous catalysis suffers from narrow applicable pH range, high reagent consumption and high risk of secondary pollution. In contrast, supported heterogeneous catalysts have become a research hotspot for BEF catalytic materials owing to their advantages including stable active sites, low metal leaching, recyclability and strong adaptability under neutral conditions. At present, BEF technology still faces challenges such as easy electrode passivation, weak anti-interference capacity in complex water matrices, high material fabrication cost, and ambiguous interfacial mechanisms underlying multi-mechanism coupling. Future research should further develop long-term stable, low-cost functional electrodes and composite catalytic materials, deeply uncover the synergistic mechanisms among microbial metabolism, electron transfer and Fenton catalysis, and promote the transformation of bio-electro-Fenton technology from fundamental laboratory research to large-scale engineering applications for practical wastewater treatment.

Keywords

Bio-electro-Fenton, wastewater treatment, in your paperelectrode materials.

1. Introduction

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ISSN: 2710-0588

With the progression of global industrialization and continuous expansion of emerging industries, the demand for industrial water maintains a steady upward trend. According to statistics from the United Nations World Water Development Report 2025, industrial water withdrawal accounts for approximately 15%–19% of the total global freshwater withdrawal. The expansion of industrial scale leads to the continuous discharge of massive industrial wastewater. Such wastewater features complex pollutant compositions enriched with various highly toxic and refractory organic contaminants, posing long-term potential threats to the stability of aquatic ecosystems in river basins and human health. At present, global water environmental control standards are becoming increasingly stringent. Conventional wastewater treatment processes exhibit obvious deficiencies in removing persistent toxic organic pollutants from industrial wastewater, and can hardly meet the ever-tightening emission control requirements. Against this backdrop, advanced oxidation processes (AOPs) are regarded as highly promising technical routes for the treatment of refractory organic wastewater [1]. As a typical advanced oxidation technology, Fenton oxidation relies on the chain reaction between H_2O_2 and Fe^{2+} to generate strongly oxidizing hydroxyl radicals (·OH), which can oxidize and decompose refractory organic pollutants in water bodies [2]. Therefore, it has been extensively investigated in the field of water pollution control. Nevertheless, the traditional homogeneous Fenton system suffers from inherent drawbacks: it operates within a narrow effective pH range, and a large amount of iron-containing sludge is continuously produced during the reaction, which markedly increases the subsequent solid waste disposal costs and restricts its large-scale engineering application [3].Hence, it is urgent to develop novel Fenton systems with both favorable economic performance and treatment efficiency to satisfy the practical demands for pollution treatment of complex industrial wastewater.

To meet specific operational requirements while minimizing reagent consumption and iron sludge generation, continuous innovations have been proposed for Fenton technology. At the beginning of the 21st century, researchers developed electro-Fenton technology, which can continuously produce H_2O_2 through electrochemical reactions. Nevertheless, this technology suffers from limited treatment efficiency and high electricity consumption, which increases the overall treatment cost. Hence, researchers have strived to innovate energy-efficient systems. In 2009, Zhu and Ni [4] first discovered that bioelectrochemical systems could generate H_2O_2. In the presence of scrap iron, hydroxyl radicals (·OH) are produced to further oxidize and remove refractory organic compounds. This breakthrough established the linkage between Fenton technology and biotechnology for the first time. The combination of bioelectrochemical systems and Fenton processes for water treatment is defined as the BEF system. In recent years, BEF systems have attracted widespread attention in water treatment owing to their prominent advantages, including low toxicity, high treatment efficiency, mild operating conditions, clean treatment processes and energy conservation [5].

The BEF system is less susceptible to pollutants with diverse characteristics, whereas its performance is relatively unsatisfactory for real wastewater treatment. Part of the energy cannot be fully utilized during wastewater degradation in BEF systems, which may be a potential reason for its limited pollutant removal efficiency. Besides, the low utilization rate of H_2O_2 leads to the waste of in-situ generated or externally supplemented H_2O_2 in the solution. In general, the challenges restricting the development of BEF systems can be summarized into five key categories: electrochemical efficiency and energy consumption, catalytic activity and stability, pH regulation, adaptability to complex wastewater, and scalability for large-scale applications.

Currently, to improve the efficiency of BEF systems, enhance their adaptability to complex wastewater environments, and reduce economic costs, in-depth research has been conducted on electrode materials [6] and catalysts [7] by researchers. This paper comprehensively summarizes and thoroughly discusses the latest research advances of BEF systems over the

ISSN: 2710-0588

past five years. The working mechanisms of BEF systems are systematically analyzed from biological and electrochemical perspectives, and the innovations and improvements of electrode materials and catalysts are summarized, providing in-depth insights for the optimization of BEF systems. Finally, the future research directions of BEF technology are outlined, aiming to provide theoretical guidance for the extensive practical application of BEF technology.

2. Mechanism of the BEF system

The BEF system represents an integration of biological approaches, electrochemical techniques and Fenton processes. Typically, its configuration comprises an anode chamber, a cathode chamber, an external load, and either a cation exchange membrane or proton exchange membrane.

$$C_xH_yO_z + (2x - z)H_2O \rightarrow xCO_2 + (4x + y - 2z)H^+ + (4x + y - 2z)e^- \quad (1)$$

$$O_2 + 2H^+ + 2e^- \rightarrow H_2O_2 \quad (2)$$

$$H_2O_2 + Fe^{2+} + H^+ \rightarrow Fe^{3+} + \cdot OH + H_2O \quad (3)$$

$$\cdot OH + \text{pollutants} \rightarrow CO_2 + H_2O \quad (4)$$

$$O_2 + H^+ + e^- \rightarrow HOO\cdot \quad (5)$$

$$HOO\cdot + H^+ + e^- \rightarrow H_2O + O\cdot \quad (6)$$

$$O\cdot + H^+ + e^- \rightarrow HO\cdot \quad (7)$$

$$HO\cdot + H^+ + e^- \rightarrow H_2O \quad (8)$$

$$HOO\cdot + H^+ + e^- \rightarrow H_2O_2 \quad (9)$$

2.1. Anodic Reaction

In the Bio-electro-Fenton (BEF) system, the anode relies on an anaerobic bioanode as the core. Electroactive microorganisms colonize the surface of conductive electrodes to form functional biofilms, which act as biocatalysts to drive substrate oxidation reactions. Microorganisms utilize biodegradable organic carbon sources such as acetate and glucose for anaerobic metabolism and release chemical energy stored in substrates, accompanied by the generation of protons and free electrons during metabolism. Electrons are transferred to the solid anode via three extracellular electron transfer pathways: cytochrome-mediated direct electron transfer, indirect electron transfer involving redox mediators, and microbial nanowires. Subsequently, electrons migrate directionally to the cathode region through the external circuit. The produced H^+ diffuses across proton exchange membranes or cation exchange membranes into the cathode chamber, providing proton sources for the two-electron oxygen reduction

ISSN: 2710-0588

reaction to generate hydrogen peroxide at the cathode. The rate of substrate oxidation by anodic microorganisms, structural stability of biofilms, and efficiency of extracellular electron transfer jointly determine the electron output capacity of the system, which serves as the energy foundation for sustaining continuous electro-Fenton reactions at the cathode.

At present, most identified electrogenic microorganisms belong to Proteobacteria and Firmicutes, including Geobacter [8], Thiobacillus [9], Escherichia coli [10], Shewanella [11], Pseudomonas [12], Zymomonas [13], Clostridium, as well as a small number of fungi (e.g., yeasts) and archaea (e.g., methanogens). The majority of these microorganisms realize direct electron transfer. Among them, Geobacter, Thiobacillus and Shewanella achieve electron transfer via nanowires and intercellular contact, exhibiting relatively high electron transfer efficiencies [14]. Saccharomyces cerevisiae and Clostridium transfer electrons through electron mediators, while Escherichia coli relies on intercellular contact for electron transfer with relatively low electron transfer efficiency [15]. With further advances in research, more electrogenic microorganisms are expected to be discovered or cultivated, especially those capable of surviving and propagating under extreme environments [16].

The electricity-generating performance of the anode is not only affected by the extracellular electron transfer of microorganisms, but also largely depends on the properties of anode materials. Developing anode materials with large specific surface area and favorable biocompatibility can effectively improve the power generation performance of anodes.

2.2. Cathodic Reaction

In the Bio-electro-Fenton (BEF) system, the cathode serves as the core functional zone for pollutant degradation. Relying on electrons supplied from the anode, protons transported across the membrane and dissolved oxygen, the two-electron oxygen reduction reaction proceeds to in-situ generate H_2O_2, which acts as an essential prerequisite for triggering the electro-Fenton reaction. The free Fe^{2+} in the system further reacts with hydrogen peroxide via the classic Fenton reaction to produce highly oxidizing hydroxyl radicals (·OH) [17]. The generated Fe^{3+} can be continuously reduced and regenerated into Fe^{2+} with the electrons from the external circuit, sustaining the iron cycle. The ·OH can non-selectively attack refractory organic pollutant molecules, destroy organic functional groups and break carbon chain structures, and ultimately oxidize organic contaminants step-by-step until they are mineralized into carbon dioxide and water. The oxygen reduction selectivity of cathode materials is of vital importance. An ideal cathode can preferentially facilitate the $2e^-$ oxygen reduction pathway and suppress the side reaction of four-electron oxygen reduction, thereby preventing the invalid consumption of electrons.

Multiple radical quenching side reactions and competitive reduction processes coexist in the cathode region, limiting the pollutant degradation efficiency. Excessive H_2O_2 in the system can trap hydroxyl radicals, and ·OH can also undergo recombination reactions with each other, leading to the loss of reactive oxidative species. Furthermore, Fe^{3+}, dissolved oxygen and intermediate reduction products within the system compete for limited electrons. The pH condition exerts remarkable regulation on the cathode reaction process. Iron hydroxide precipitates tend to form under neutral and near-neutral conditions, blocking the Fe^{2+}/Fe^{3+} cycle and drastically lowering Fenton efficiency, whereas acidic environments are more favorable for stabilizing iron ions. Unlike the anode, which mainly provides electrons, the BEF cathode integrates the whole processes of in-situ oxidant generation, iron cycle and advanced oxidation of pollutants. The kinetics of cathode reactions, dissolved oxygen supply rate, iron ion concentration and system pH jointly govern the removal efficiency of refractory organic pollutants, accomplishing the ultimate pollutant conversion step in the coupled system of bioelectron generation and advanced oxidation.

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3. Electrode Material

As can be concluded from the above mechanism analysis, electrode materials play a crucial role in BEF systems. Researchers have carried out comprehensive investigations to enlarge the surface area and improve the biocompatibility of anode materials, as well as optimize the selectivity of cathode materials toward the two-electron oxygen reduction reaction.

3.1. Anode Material

In the Bio-electro-Fenton system, the anode serves as the core unit for electron supply. The physicochemical properties of electrode materials directly govern the colonization of electroactive microorganisms, biofilm development and extracellular electron transfer (EET) efficiency, and ultimately regulate the sustainability of the cathodic Fenton reaction. Traditional anodes for BEF are mainly based on pristine carbon materials including carbon felt, carbon cloth, graphite brush and granular activated carbon [18]. These materials possess favorable electrical conductivity, chemical stability and biocompatibility, making them the most widely used substrates in laboratory research at present. Nevertheless, pristine carbon materials generally suffer from limited specific surface area, insufficient surface active sites and high interfacial charge transfer resistance, which easily lead to inadequate biofilm loading and hindered electron transfer inside thick biofilms. Modification strategies for pristine carbon materials have been extensively investigated. The mainstream approaches include surface oxidative etching, heteroatom (N, O, P) doping, and loading nanocarbon materials such as carbon nanotubes and graphene. By increasing oxygen-containing functional groups, constructing hierarchical porous structures and optimizing electrode hydrophilicity, these modifications provide more attachment sites for electrogenic microorganisms and strengthen the cytochrome-mediated direct electron transfer pathway. In recent years, biomass-derived biochar anodes fabricated via pyrolysis of agricultural and forestry wastes and organic solid wastes have developed rapidly. Benefiting from low-cost raw materials and solid waste recycling, combined with pore structure and surface functional group regulation through activation treatment, biomass biochar has gradually become an important research direction for low-cost anode materials.

To improve the specific surface area of bioanodes, researchers have developed three-dimensional carbon-based materials such as carbon nanotubes and carbon nanofibers. These materials feature abundant porous structures and low internal resistance, and facilitate the formation of three-dimensional biofilms, thereby boosting mass transport and electron transfer. Consequently, they can enhance the pollutant removal efficiency of BEF systems. For instance, Heidari et al. constructed a three-dimensional electrode using granular activated carbon in the anode chamber of a BEF system. After 72 h, the removal efficiencies of methyl orange in the cathode chamber (73.56% decolorization efficiency and 69.76% chemical oxygen demand (COD) removal efficiency) exceeded those of the two-dimensional electrode system (70.75% decolorization efficiency and 64.82% COD removal efficiency).

Furthermore, scholars have modified electrodes to strengthen the attachment of electrogenic microorganisms on electrodes and accelerate electron transfer, so as to elevate the power generation performance of bioanodes. Common modification strategies include polymer deposition, oxygen or nitrogen doping, immobilization of electron mediators, and incorporation of carbon nanoparticles. Huang et al. modified carbon fibers with vertically aligned carbon nanotube/polyaniline composites to improve electrical conductivity and biocompatibility. The power output was increased by 2.63 times compared with the unmodified anode. Such performance improvement can be mainly attributed to the enlarged specific surface area brought by vertically grown nanotubes, as well as the enhanced surface biocompatibility originating from the polyaniline coating [19].

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3.2. Cathode Material

In BEF systems, cathode materials act as electron donors or catalytic surfaces during electrochemical reactions to facilitate the reduction of oxygen to H_2O_2. These materials occupy a central position in electrochemical processes and determine the electron transfer efficiency as well as the oxygen reduction reaction (ORR) performance of the system. Conventional cathode materials are mainly common carbon-based materials such as graphite, carbon cloth and carbon felt. Although these materials feature easy availability and strong chemical stability, pristine carbon materials exhibit poor selectivity toward the $2e^-$ ORR and yield low H_2O_2 production. To optimize catalytic performance, numerous modification strategies have been continuously developed. Acid-base activation and plasma etching can be adopted to regulate surface defects and oxygen-containing functional groups of carbon materials; heteroatom doping (N, O, F, etc.) can tune the electronic structure of carbon and optimize the energy barrier for oxygen adsorption and activation. Meanwhile, composite modification of cathodes with nanocarbon materials including carbon nanotubes, graphene and porous biochar has been extensively investigated.

Moreover, loading iron/cobalt-based single atoms, metal oxides, and metal-organic framework (MOF)-derived carbon catalysts on carbon substrates can effectively enhance the catalytic selectivity for the $2e^-$ ORR and boost H_2O_2 accumulation in the system. Some catalysts can simultaneously catalyze the reduction of Fe^{3+} and accelerate the internal iron ion cycle within the system. In addition, air diffusion electrodes have been developed and applied to reduce aeration energy consumption. Such electrodes allow sufficient gas to penetrate into the electrode interior and interact with electrolyte solutions to construct a three-phase gas-liquid-solid interface for catalytic reactions.

Ren et al [20]. fabricated an innovative air cathode using thermally treated candle soot. Thermally treated candle soot possesses a large specific surface area, abundant mesoporous structures, favorable hydrophilicity and abundant oxygen-containing functional groups, all of which contribute to H_2O_2 generation during oxygen reduction. The air cathode derived from thermally treated candle soot achieves a high selectivity of 95.0%–97.5% toward the two-electron ORR, and its H_2O_2 production rate is 6.29 times that of the carbon black air cathode.

4. Catalyst

4.1. Homogeneous Catalyst

Homogeneous catalysis represents a core strategy to construct efficient Fenton chain reactions in Bio-electro-Fenton (BEF) systems. Catalysts are homogeneously dispersed in the cathodic electrolyte in the form of soluble ions, which enables sufficient contact with in-situ generated H_2O_2 on electrodes. This approach effectively addresses the drawbacks of solid catalysts, including large mass transfer resistance and insufficient exposure of active sites. At present, the Fe^{2+}/Fe^{3+} iron-based homogeneous catalytic system is the most widely adopted catalyst in BEF systems. In 2009, Zhu and Ni first introduced this system into microbial electro-Fenton systems and realized efficient degradation of p-nitrophenol, verifying the feasibility of combining homogeneous catalysis with bioelectrochemistry for the degradation of refractory pollutants. Existing studies have demonstrated that optimizing Fe^{2+} dosage can increase the removal efficiency of methyl orange dye in BEF systems from 73.9% to 86.7%, demonstrating excellent catalytic activation performance. Benefiting from the coupling of electron supply from the anode and in-situ H_2O_2 generation at the cathode, stable Fe^{2+}/Fe^{3+} cycling and continuous production of hydroxyl radicals (·OH) can be achieved for the efficient degradation of phenolic and tetracycline organic pollutants.

Nevertheless, traditional iron-based homogeneous catalysis suffers from prominent limitations. High catalytic activity can only be maintained under strongly acidic conditions at pH 2.5–3.5.

ISSN: 2710-0588

Iron ions tend to undergo hydrolysis and form hydroxide precipitates under neutral conditions, leading to catalyst deactivation. Moreover, excessive iron ions trigger radical quenching side reactions and weaken the mineralization of pollutants. To broaden the applicable pH window, researchers have modified iron ions with complexing agents such as oxalic acid, citric acid, humic acid and EDTA. The formation of soluble coordination complexes inhibits iron precipitate formation. Among these candidates, the iron-oxalate system can effectively extend the efficient operating range of BEF systems to neutral environments. Meanwhile, multimetal homogeneous catalytic systems such as Cu^{2+}/Cu^{+} and Co^{2+}/Co^{3+} have been gradually explored. Their distinctive redox potentials can compensate for the performance deficiencies of single iron-based catalysts.

In addition, Wang et al [21].utilized ammonium nitrogen in wastewater as electron donors. Iron-reducing bacteria immobilized on biocathodes were applied to regenerate ferrous ions from iron sludge. The application of iron-reducing bacteria in BEF systems offers a new strategy to overcome the inherent limitations of homogeneous BEF reactions and provides guidance for future research on BEF technology.

4.2. Heterogeneous Catalyst

Heterogeneous catalytic Bio-electro-Fenton systems have attracted extensive research interest in the BEF field in recent years owing to merits including easy solid-liquid separation, recyclability, low risk of secondary pollution and a wider applicable pH range [22]. Different from free metal ions, heterogeneous catalysts exist in solid form with active sites distributed on the surface of solid supports. Common supports consist of carbon materials, clay minerals, zeolites, and metal-organic framework-derived carbon. The dominant active components involve zero-valent iron, iron oxides, layered double hydroxides (LDHs), and iron-based single-atom materials. For example, a composite cathode catalytic system constructed by immobilizing Fe_3O_4 on carbon felt enables continuous degradation of tetracycline under near-neutral conditions, and the catalyst retains stable catalytic activity after multiple cycles [23]. When LDH-derived Fe-Co bimetallic composite catalysts are applied in BEF systems, the bimetallic synergistic effect accelerates Fe^{3+} reduction, effectively facilitating the Fenton cycle and improving the mineralization efficiency of Rhodamine B [24]. Meanwhile, numerous studies have focused on MOF-derived carbon-based heterogeneous catalysts. The abundant pore structures of such materials enhance the adsorption and activation capacity toward H_2O_2 and alleviate interfacial mass transfer limitations [25]. In addition, researchers have developed composite iron-based nanocatalysts, such as Fe-MOFs, Fe@Co composites and Fe@S nanoparticles (NPs). Among these iron-based nanomaterials, maghemite nanomaterials derived from magnetite can be readily recovered with magnetic fields. Nevertheless, the synthesis of magnetite nanomaterials generally consumes massive raw materials, involves complicated procedures and requires high electric energy input, which raises catalyst costs. Zhao et al. proposed a novel strategy to extract magnetite components from fly ash for fabricating nano-sized Fe_3O_4 catalysts, thereby cutting raw material costs [26].

To tackle the above challenges of heterogeneous catalysts, researchers immobilize catalysts onto cathodes to prepare supported heterogeneous catalysts. This strategy not only realizes in-situ generation and decomposition of H_2O_2, but also avoids iron ion leaching and reduces subsequent treatment costs. At present, typical supported heterogeneous catalysts include iron oxide-supported catalysts, supported iron-based metal-organic frameworks (MOFs), zero-valent iron-supported catalysts and iron sulfide-supported catalysts. However, supported catalysts still face certain drawbacks. High loading dosage easily causes agglomeration of active particles and pore blockage, whereas low loading dosage leads to insufficient active sites. Moreover, organic intermediates tend to cover catalytic interfaces during long-term operation and induce electrode passivation, resulting in the decay of pollutant degradation performance

ISSN: 2710-0588

[27]. Overall, benefiting from prominent advantages such as controllable structure, strong stability, electrode integration capability and wide pH adaptability, supported heterogeneous catalysts have become the mainstream development trend of BEF advanced oxidation technology. Future research should further optimize loading procedures, strengthen interfacial bonding strength, and construct anti-fouling long-lasting catalytic systems, so as to promote the large-scale application of supported composite catalytic electrodes in the treatment of actual complex wastewater [28].

5. Conclusion

In summary, the Bio-electro-Fenton (BEF) system integrates the merits of bioelectrochemical power generation and Fenton advanced oxidation, and achieves low-carbon and efficient treatment of refractory organic wastewater via coupled anodic and cathodic reactions. The bioanode oxidizes organic substrates in water through anaerobic metabolism of electroactive microorganisms and continuously outputs electrons and protons, providing stable charge supply for the electrocatalytic reactions of the system. At the cathode, hydrogen peroxide is generated in situ through the two-electron oxygen reduction pathway. Combined with iron-based catalytic systems, a redox cycle is established to produce strongly oxidizing hydroxyl radicals, thereby realizing efficient mineralization and degradation of organic pollutants. Current optimization research on BEF wastewater treatment technology mainly focuses on functional modification of electrodes and upgrading of catalytic systems. Structural regulation, heteroatom doping and nanocomposite modification of carbon-based materials can effectively improve the electron transfer efficiency and catalytic selectivity of electrodes and enhance the overall reaction kinetics of the system. For catalytic systems, traditional homogeneous iron-based catalysis exhibits high reactivity, yet suffers from poor pH adaptability, high reagent consumption and risks of secondary pollution. In contrast, supported heterogeneous catalytic materials feature outstanding stability, recyclability, low metal leaching and favorable performance under neutral conditions. Relying on the interfacial synergistic effect between supports and active components to optimize catalytic cycles, such materials have become the core research focus in the BEF field.

BEF technology effectively overcomes the limitation of conventional biological methods in treating highly toxic and refractory organic wastewater, and thus possesses promising application and development prospects in wastewater treatment. Nevertheless, obvious bottlenecks still hinder its practical application. Electrodes are prone to passivation induced by the coverage of degradation intermediates during long-term operation, leading to gradual deterioration of system performance. Impurities contained in complex real water matrices interfere with electrode reactions and catalytic processes, and the system exhibits limited anti-interference capacity. In addition, the fabrication of high-performance modified electrodes and catalysts involves high costs and complicated procedures. Furthermore, the in-depth multi-field coupling mechanisms among microorganisms, electrochemistry and catalysis have not been fully clarified, which restricts the industrial implementation of this technology.

Future research should prioritize the development of long-term stable and low-cost modified electrodes and supported catalytic materials. It is essential to deeply reveal the coupled mechanisms of interfacial electron transfer, iron cycle regulation and pollutant degradation, and improve the anti-fouling and anti-interference capacity of the system. These efforts will facilitate the transition of BEF technology from laboratory-scale trials toward large-scale engineering applications for complex industrial wastewater and municipal sewage.

ISSN: 2710-0588

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