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Electric biological coupling process (EBCP) for wastewater treatment: a general review

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ABSTRACT

It is well known that individual classical wastewater treatment techniques (e.g. activated sludge process, anodic oxidation, etc.) are generally inadequate for the removal of hazardous substances from integrated urban wastewater. Therefore, electric biological coupling process (EBCP) has been proposed in order to ensure the effective removal of these pollutants, as each of its constituent techniques supplements the advantages and overcomes the challenges of the other. This paper presents a comprehensive review of the development and application of this process, summarizing the concept, design and operating principle of EBCP system. Various electrode materials used in an EBCP reactor are systematically introduced, including anode and cathode materials as well as particle electrode materials. The effects of important operating parameters, such as current density, cell voltage, dissolved oxygen concentration, pH values and influent loading are discussed. Their applications in treating various types of wastewater are thoroughly reviewed. Finally, perspectives are also proposed on the future trends in the development of the technology. This long-expected overview aims to draw attention to the innovative process, promote its application and development as well as bring the scientific community up-to-date with the latest advances in the field.

Keywords: Electrochemistry; Biological treatment; Electric biological coupling technology; Coupling mechanism; Electrode material; Space structure

1. Introduction

Heavy metals, emerging organic contaminants (EOCs) and other non-biodegradable pollutants from domestic, commercial, and industrial sources have been widely found in sewage, surface, ground and drinking water samples in different countries [1–3]. Environmental pollution by these hazardous substances poses a severe ecological problem. This is further complicated by the fact that most of them are difficult to degrade using conventional wastewater treatment methods [4]. A significant number of works deal with the methods of handling these toxic pollutants, mainly in full-scale wastewater treatment plants (WWTPs) based on conventional activated sludge (CAS) processes. However, conventional wastewater treatment processes have a number of disadvantages. Firstly, they do not always effectively remove toxic substances from wastewater, potentially bringing secondary pollution to receiving waters [5–8]. This affects in turn the quality of the water source of supply plants and local groundwater. Secondly, for many WWTPs, sewage sludge disposal is perhaps one of the most pressing problems, since conventional wastewater treatment of sludge has obvious limitations [9]. Thirdly, conventional supply water treatment processes, such as coagulation, precipitation, or filtration fail to achieve satisfactory performance for these pollutants [10,11]. Advanced oxidation process (AOP) is a commonly used water treatment technology, which has become

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extremely popular as a method of organic pollutant treatment thanks to its ability to achieve total mineralization due to the formation of hydroxyl radicals (•OH) [12]. AOP involves pollutant oxidation by: (i) direct electron transfer to the anode surface M, (ii) heterogeneous reactive oxygen species (ROS) produced as intermediates of oxidation of water to oxygen, including the powerful •OH physisorbed at the anode surface, denoted as M(•OH), generated via Eq. (1), and weaker oxidants such as H_2O_2 produced from M(•OH) dimerization, as shown in Eq. (2) [13–15]. However, there are still some factors limiting their application such as a high level of energy consumption as well as considerable construction and maintenance costs. Thus, finding a way to remove these substances in an affordable and efficient manner is an urgent challenge.

$$M + H_2O \rightarrow M(\bullet OH) + H^+ + e^- \quad (1)$$

$$2M(\bullet OH) \rightarrow 2MO + H_2O_2 \quad (2)$$

Nowadays, bio-chemical and electrochemical technologies are the two main methods of advanced tertiary treatment. Bio-chemical technologies remove smell, color and organic substances. Their advantages include mature technology, convenient management, low operating costs and ease of operation [16,17]. In spite of these advantages, some shortcomings do occur, e.g. incomplete degradation of toxic emerging organic pollutants, slow decomposition rate, and sometimes microorganism inactivity due to poisoning [18]. Electrochemical processes such as electrochemical conversion and electrochemical combustion remove contaminants mainly by redox reaction and deliver higher removal efficiency, contributing to improved biodegradability (BOD_5/CODcr)[19]. Electrochemical technologies have made a great step forward in wastewater treatment, especially for bio-refractory substance abatement due to their high efficiency, environmental friendliness and versatility [20]. However, side effects are numerous, including oxygen, chlorine and hydrogen emission as a by-product, lower current efficiency, temperature increase during the process, high operating cost and high energy consumption [21,22]. To supplement the characteristics and compensate for the limitations of these two water treatment technologies, a new electric biological coupling technology (EBCP) has been proposed. This approach not only combines the advantages of the constituent treatment techniques but also eliminates the drawbacks of each of them. The global spread of EBCP is slow, but it has drawn wide attention in the wastewater treatment in recent years. Its application involves removing metal ions [23] at the early stage from different types of wastewater, including nitrate wastewater [24], phosphorus wastewater [25], dyestuffs wastewater [26], nitrobenzene wastewater[27], toxic and refractory organic wastewater [28] and some other pollutants. Unfortunately, to the best of our knowledge, no comprehensive reports are available on EBCP's performance in wastewater treatment [29,30].

This paper presents a general review of EBCP, including its concept, design and operating principle, while also focusing on electrode selection, operating parameters, reactor structure, and its application for treating various wastewaters. At the same time, perspectives on the future trends in the development of the technology are also proposed. It is expected that the review will attract more attention to this promising technology and contribute to its development.

2. Electric biological coupling process (EBCP)

2.1. Definition and development

EBCP is a process whereby electrochemical reactions and microbial reactions are coupled in a single reactor implementing autotrophic denitrification, simultaneous nitrification and denitrification, and degradation of organic substances, aiding in the removal of biological phosphorus and heavy metal ions. EBCP ensures higher removal efficiency and reduced engineering costs.

In 1988, Fuchs et al. [16] were the first to combine biological treatment with electrochemical technology for denitrification. Mellor et al. [31] pioneered the concept of current promotion, electrode bioreactor and denitrification control. Afterwards, EBCP was widely used to treat different types of wastewater, such as toxic and refractory organic wastewater as well as wastewater containing heavy metal ions. EBCP shows a certain superiority compared to conventional wastewater treatment processes. Unfortunately, the development of EBCP is still at an early stage internationally. The factors mainly taken into account in pollutant removal are electric field strength [32,33], pH, temperature [34] and electrode materials [35,36]. However, little is known about the action of microorganisms with electric fields, whereas the underlying theory is inadequate [37].

2.2. The structure of EBCP

EBCP is an organic combination of electrochemical reaction and biological treatment, whose structure should be consistent with electrode and microbial needs. The rate of degradation or decomposition is mainly determined by the structure of EBCP, which if designed properly, may establish an efficient relationship between conductance and mass transfer, improve the reaction dynamics of the coupling system, shorten reaction times, save land and facilitate the removal of pollutants. As regards the form of coupling of electrodes and microbes, it is possible to distinguish three main types of reactors within the EBCP, namely a two-dimensional electro-bioreactor (Fig. 1a, Fig. 1b), three-dimensional electro-bioreactor (Fig. 1c) and composite electro-bioreactor (Fig. 1d).

2.2.1. Two-dimension electro-bioreactor

Two-dimensional electro-bioreactor represents a technology in wastewater treatment using pure or mixed microorganisms as biofilm attached to the electrodes [38,39]. In these systems, oxidative and reductive reactions are catalyzed by biofilm-electrodes [40,41]. Applying an electric field accelerates the ion migration rate, enhancing the reactions taking place on the electrode surfaces. Usually, Two-dimensional electro-bioreactor is seeded with activated sludge from a municipal waste-

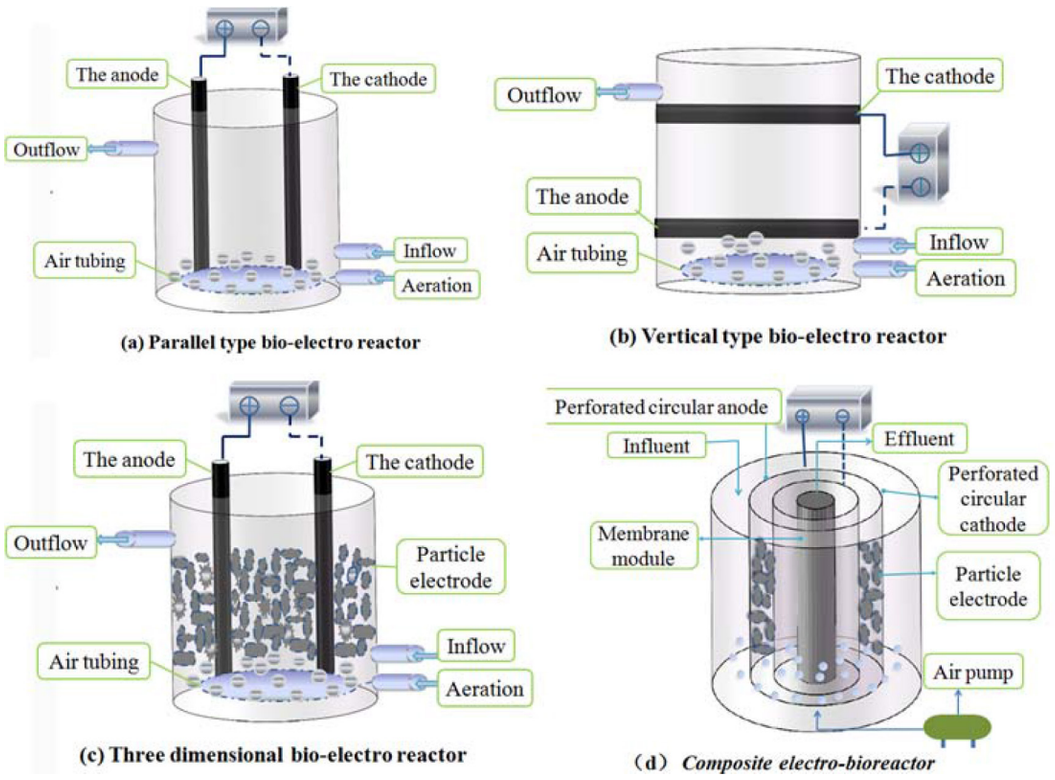

Fig. 1. Some typical structure of EBCP.

water treatment plant through the action of the applied electric field. The electric field promotes the coagulation of colloidal substances by flocs, providing a large surface area for sorption. Hoseinzadeh et al. [42] showed that the coupling of low-voltage, low-frequency alternating electric current and biofilm in an anoxic reactor leads to the generation of dense and fast-settling sludge granules. Liu et al. [43] suggested that the use of a zerovalent iron combined with an electric field in an upflow anaerobic sludge blanket reactor could effectively enhance sludge granulation. Accordingly, the relationship between current density and the direction of wastewater flow can be divided into parallel (Fig. 1a) and vertical types (Fig. 1b). A parallel reactor is one in which the current direction is parallel to wastewater flow, and electrode can be plate or strip-shaped. The possibilities and capabilities of a parallel reactor are widely known, but there is a conflict between the uniformity of electric potential and conversion rate. In order to improve the conversion rate, a considerable length of time is necessary, but the electric potential distribution can be uniform only when the current path is short. On the other hand, when the current direction and wastewater flow are vertical to each other, the wastewater in the reactor has sufficient residence time, and electric potential distribution is uniform. This means that the vertical reactor overcomes these shortcomings, providing the efficient removal rate for pollutions. Kuroda et al. [44] investigated the application of electro-bioreactor process for the treatment of wastewater containing nitrate and internal source of hydrogen donor as organic matter. It was confirmed that COD as well as nitrate were removed simultaneously by applied electric current in a continuous experiment.

2.2.2. Three-dimensional bio-electro reactor

It is well known that three dimensional (3D) electrochemical process is established based on two-dimensional electrochemical process which is largely similar in terms of electrode materials and treatment procedures, except with the addition of a third electrode. Also known as the particle electrode or bed electrode, it is basically comprised of granular or fragmental materials placed between two counter-electrodes. Simultaneously, the microorganism is attached to particle electrode and main electrode. Under optimized treatment conditions, microorganisms can effectively degrade pollutants through catabolism and electrochemical action. Three-dimensional electrode technology and biological treatment technology are combined to form a three-dimensional electrode biofilm reactor. In such processes, with removal involving adsorption, water electrolysis, microbial degradation, and electro-oxidation processes [45], microorganisms use conductive particle electrode as channel for electrons, ensuring efficient interspecies electron transfer and cooperative catabolism during electrically supported microbial growth. This produces higher specific surface area and shorter distance of mass transfer, contributing to better effectiveness, especially for applications with

wastewater having low reaction rate. Mascia et al. [46] proposed a packed bed electrochemical reactor, with 3D electrodes of conductive diamond used to investigate the treatment of water containing M. aeruginosa. Based on the experimental results, the maximum values of algae inactivation were obtained at R_e (Schmidt number) = 10 and i (current density) = 25 A/m^2.

2.2.3. Composite electro-bioreactor

A composite electro-bioreactor generally refers to several wastewater treatment processes coupled to a hybrid reactor. It is a combination of biological, electrokinetic and membrane filtration or other wastewater treatment processes in one hybrid reactor. For example, submerged membrane electro-bioreactor (SMEBR) is a new synthetic technology for wastewater treatment applying electrical field and microfiltration in a nutrient-removing activated sludge process [47]. In this process, the removal of pollutants is mainly attributed to biodegradation, electrocoagulation and membrane filtration. Gao et al. [48] reported submerged membrane electro biology reactor (SMEBR) which is an improved version of biofilm reactor (MBR). SMEBR can eliminate 80–100% of chemical oxygen demand (CODcr). Compared with the traditional film technology, the removal rate of total nitrogen in the wastewater can be increased by 30%, with removal rates for soluble phosphate above 99% under low-carbon conditions. Yu et al. [49] reported that graphite electrode was used in an anaerobic and aerobic fluidized bed to construct an electrochemical A/O bio-fluidized bed for the treatment of coking wastewater. Liu et al. [50] introduced an electrode biofilm process into the sequencing batch biofilm reactor (SBBR) for the denitrification of municipal wastewater. Under the best operating conditions, the highest removal rate of NO_3^--N was up to 89.48% and the removal rate of total nitrogen was 66.67–84.37% over a period of 495 min. Feng et al. [51] constructed a TDE-BAF reactor, mainly using a combination of three-dimensional electrode technology (TDE) and biological aerated filter (BAF). TDE-BAF not only combines the advantages of BAF's low cost and TDE's effective degradation of organic substances, but also allows using the side reactions, which occur in TDE, for biological reactions to improve current efficiency and pollutants removal rate.

2.3. Mechanism and characteristics

It is well recognized that EBCP is established based on electrochemical process and biological treatment technology with many similarities such as electrode materials and treatment processes. Biodegradation is the process whereby organic pollutants with large molecular weight are degraded into small molecules by microorganisms such as bacteria, algae and fungi [52], and even bio-mineralised to simple inorganic molecules such as water and carbon dioxide. In conventional biodegradation process, microorganisms use organic compounds as primary substrates for cell growth and produce enzymes for their assimilation [53]. However, the EBCP mechanism is complex, featuring an electric field and microorganisms in a single reactor. In simple terms, the EBCP involves bacteria exchanging electrons with the electrodes to treat wastewater [54]. Under the action of applied electric field, active substances, which are supposed to be generated from electrochemical processes, accelerate the metabolic activity of microorganisms. The processes and mechanisms of electron transfer through microorganisms and microbial redox have been widely investigated. For example, recent studies [54,55] have reported that some bacteria perform anodic and cathodic half-reactions using electrodes as electron donors and acceptors, respectively. Three types of interspecies electron transfer (IET) through a microbial community have been described. These include: (a) diffusion of redox chemical species [56], (b) direct contact with electrodes in cell aggregates [57], and (c) natural conductive minerals (oxides of iron-magnetite) and/or direct contact in cell aggregates. Li et al. [58] reported that photoheterotrophic microorganisms catalyze the anode reaction and provide oxygen as an electron acceptor to the cathode reaction. If particle electrode is present in EBCP, microbes use conductive mineral particles as channels of electrons, resulting in efficient IET and cooperative catabolism during electrically supported microbial growth. Therefore, in EBCP, many contaminants are efficiently removed by the electrocatalytic redox reaction, biological metabolism, physical adsorption and retention by filler. The four main characteristics of EBCP are as follows:

1) The electrochemical reaction and biological treatment are combined in a single reactor, providing the additional advantages of compact equipment, small footprint, simple operation and management;
2) It can simultaneously play the role of electrocatalytic redox reaction, bio-oxidation and physical adsorption;
3) It also can weaken the impact of toxic substances on microorganisms, resulting in a high removal efficiency and good water quality; as a result, it offers wide opportunities for use in industrial wastewater degradation and advanced treatment of urban sewage;
4) More importantly, the electron transfer process is dependent on the distance between redox center and electrode [59]. EBCP shortens the distance of mass transfer by direct contact of microorganisms with electrode [60], so it is very suitable for engineering reconstruction and expanding for WWTPs.

3. Course of research on EBCP

3.1. Electrode materials

In EBCP, electrode material (anode, cathode and particle electrode) plays an important role in electron transfer and may be involved in electrode reaction. The materials of anode, cathode and particle electrodes applied in EBCP have been summarized in Table 1. In the process of electron transfer, the so-called redox mediators are electrochemical active substances which can participate in the microbial respiration process and accept the electrons, e.g. oxygen, from the electron transport chain (ETC) [61,62]. Therefore, electrode material is one of the most important influence factors determining the efficiency, capacity,

service life and cost of the process. In an excellent EBCP system, the cathode and anode materials should have good electrocatalytic performance, long service life and high stability. As far as we know, a preferable particle electrode requires properties including higher specific surface areas, outstanding catalytic activity, adequate mechanical strength and conduciveness to microbial attachment growth. Recently, numerous studies have been focusing on the preparation of particle electrodes with high activity and stability [63]. Particle electrodes frequently used in three-dimensional electrode electrochemical reactions are predominantly made of carbonaceous [64] and metallic (including metal oxide) material.

In general, the selection of electrodes in EBCP is determined by the wastewater's characteristics and the role of electrode materials, i.e. whether they are suitable for electric flocculation reduction, oxidation reduction or denitrification reduction. Some typical electrode materials used in EBCP, their characteristics and roles are summarized in Table 1. Based on a comparison between BDD (boron-doped diamond) and Pt anodes, some studies have discovered that BDD exhibits superiority over Pt, both in terms of decolorization and mineralization [65–67], since BDD shows a much higher potential for O_2 evolution, producing larger amounts of $^{\bullet}OH$ to degrade organics [68–71]. Generally speaking, when electrocoagulation takes place in aqueous solution, the material usually chosen for the anode is cheap and corrosive, such as aluminum or iron, while the cathode is fabricated to resist corrosion. If the reactor features mainly oxidation and reduction in EBCP, the anode material should be characterized by high oxygen potential and catalytic performance, while the cathode is usually made of stainless steel. In order to remove nitrate by denitrification, it is vital to choose an anode with high oxygen potential material and a cathode with activated carbon fiber (ACF). Some types of particle electrodes are used in EBCP, including activated carbon, metal particles and supported metals or metal oxides particles. However, these particle electrodes have the following problems:

1. Activated carbon particles may easily generate short-circuit current [72] because of small resistance and are prone to cause pulverization.
2. The metal oxide particle electrode process will produce some toxic ions in the electrolysis, resulting in the formation of secondary pollutants, such as PbO_2 particle electrode.
3. The particle electrode with metal or metal oxide overcomes the shortcomings of activated carbon particle electrode and improves the effect of sewage treatment, but is problematic in that loading metal or metal oxide may easily fall off [73–75].

3.2. Influence of operating parameters

EBCP is a complex multi-enzyme system. Its efficiency is affected by numerous factors which also impact on the growth of microorganisms, the mode of electric field, the mass transfer of the reaction kinetics, reactor structure etc. The main impact factors are summarized below.

3.2.1. Effect of current density or voltage

Current density and voltage are among the most important variable parameters in EBCP since they not only affect the electrochemical oxidation and capital cost [87], but also impact the metabolism of microorganisms. When current density or voltage is used as a controlling parameter, the processing power increases correspondingly with the rise in current density or voltage. However, treatment performance does not always improve accordingly accompanying with the increase of energy consumption (EC). For example, high chloramphenicol (CAP) degradation efficiency was not obtained at high output power. The EC of degrading CAP was 34.16 kWh/kg CAP at current intensity of 0.05 A. However, when the current intensity increased to I = 0.5 A, the EC was 765.96 kWh/kg CAP, which was an increase of more than 2200%. The results showed that excessive current intensity could reduce CAP degradation efficiency [88]. More importantly, an excessively high voltage may induce microbial metabolic disorders, or even kill microorganisms [89]. Numerous experiments indicate that a further increase in current density or voltage also did not considerably improve COD or color removal for many wastewaters [90,91]. Wei et al. [92] studied the effects of different current densities on the number, species and treatment capacity of microorganisms. When current density was less than 6.2 A/m^2, the viability of microorganisms was not significantly affected and the mortality was less than 10% after 4-hour treatment. At current densities of 12.3 A/m^2 and 24.7 A/ m^2, the number of viable cells decreased by 15% and 29%, respectively. Zeyoudi et al. [93] studied the impact of continuous and intermittent supply of electric field to microbial community used for wastewater treatment. They found that continuously applied electric current density has no effect on microbial population structure in a wastewater bioreactor. Constant low electric current densities stimulate the growth of microorganisms and this effect seems to be nonselective. Also, intermittently applied electric current density causes a shift in the microbial population structure in a wastewater bioreactor. Intermittent electric current density either stimulates the growth of a subset population or selectively eliminates or inhibits the growth of a subset population of microorganisms in a bioreactor.

3.2.2. Effect of dissolved oxygen or gas-water ratio

During EBCP, the compressed air was uniformly sparged into the electro-bioreactor by a micro-pore plate. The air sprayed into the EBCP has four effects: (1) aeration provides oxygen for the microorganisms; (2) aeration may play a role in stirring, thereby strengthening the mass transfer efficiency and eliminating the concentration polarization phenomenon [74]; (3) aeration supplies essential oxygen. Under acidic conditions, the oxygen at the cathode can react with H^+ to generate H_2O_2 and then produce hydroxyl radical to oxidize [94]; (4) oxygen can reduce the side reaction of the anodic oxygen evolution to improve the current efficiency [95]. Wu et al. [95] reported that the oxygen can be changed into a stronger oxidizing agent-H_2O_2 in a three-dimensional electrode system. The

Table 1
The materials of anode, cathode and particle electrodes applied in EBCP

Electrode	Material	Properties	Application	References
Cathode	Iron sheet	Reduction and electric flocculation; Surface area: 0.5 m^2;	Azo dyes	[76]
	IrO_2 /Ti	Higher oxygen evolution potential; Size: 200 mm × 200 mm;	Penicillin kettle waste	[77]
	Porous TiO_2 film plate	Electric catalytic oxidation; Porous and specific surface area; Size: 60 mm × 40 mm;	Rhodamine B	[78]
	Ruthenium iridium titanium	Electric–catalytic; Size: 150 mm × 150 mm;	Phenol waste	[79]
	Ti–base coating	Size: 110 mm × 50 mm;	Nitrobenzene	[80]
	Sb–doped SbO_2/Ti	Catalysis; Size: 40 mm × 60 mm;	Reactive brilliant red X–3b	[81]
	Rare earth doped Ti/Sb–SnO_2	Good catalytic activity; large surface area; Size: 50 mm × 30 mm;	Phenol	[82]
	Ruthenium–plated titanium mesh	Direct and indirect electrochemical oxidation	Methyl orange	[83]
	Ti/Co/SnO_2–Sb_2O_5	Size: 60 mm × 110 mm;	Papermaking wastewater	[84]
Anode	Graphite carbon fiber; Graphite rod;	Large specific surface area; Porosity; Graphite carbon fiber easier to fall off; Size: 120 mm × 250 mm;	Sulfate	[20]
	Tin–Copper electrode; Nickel–Phosphorus electrode	Good catalytic activity; It works well in acidic and neutral conditions; Surface area: 1 cm^2	Nitrobenzene	[85]
	Rare earth metal electrode	Good catalytic activity; Easy dissolve in acidic conditions; Surface area: 4 cm^2;	Nitrobenzene	[85]
	Porous graphite electrode	Large specific surface area; Size: 150 cm × 150 cm;	Phenol waste	[79]
	Titanium plate	Size: 40 mm × 60 mm;	Reactive red	[81]
	Graphite – polytetrafluoroethylene	Electric fenton; Size: 50 mm × 30 mm;	Fenton	[82]
Particle electrode	Quartz Microporous Porcelain ring particles loaded with Sb–SnO_2	Adsorption–electrocatalytic reaction; Stable structure;	Herbicide wastewater	[86]
	Fe–Ni–TiO_2 /AC particle electrode	Synergistic catalysis; direct oxidation, hole oxidation, active material oxidation; visible light photo–sensitization degradation; Particle diameter : 3–5 mm;	Rhodamine	[78]
	Activated carbon particles	Adsorption and catalysis; Specific surface area: 1028 m^2/g; Particle diameter: 0.2 mm;	Phenol waste	[79]
	Metal composite electrode	Electric fenton;	Nitrobenzene	[80]
	Loading Sb–SnO_2 porous ceramic particles	Indirect oxidation; Improving mass transfer efficiency;	Reactive red	[81]
	Chitosan organic polymer	Good adsorption; High electron transfer rate; Excellent Photocatalytic performance;	Phenols	[82]
	Activated carbon–loaded manganese oxides, zinc oxides, iron oxides	More hydroxyl radicals; High energy consumption;	Methyl orange	[83]

gas-water ratio in EBCP determines whether the whole reactor is anaerobic, anoxic or aerobic. In an aerobic treatment process, oxygen demand is the main indicator of process control. Xiong et al. [96] observed that COD removal efficiency progressively decreased from 94.5% to 84.4% for an airflow of 7.0 L min^{-1}, while COD removal efficiency dropped from 92.7% to 68.9% in the absence of airflow. In general, when dissolved oxygen concentration is less than 2 mg/L, dissolved oxygen is the limiting factor for nitrification [97]. Meanwhile, sufficient oxygen is needed to generate the active intermediates in EBCP. Furthermore, in order to counteract the inhibiting action of dissolved oxygen, the gas-water ratio should be appropriate to control and maintain the dissolved oxygen concentration of 3 mg/L or above. However, under different operating conditions, the reaction has different optimum dissolved oxygen concentration. Smaller dissolved oxygen concentrations do not meet the basic conditions required for the reaction, but a larger airflow can easily wash off the microorganism attached to the particle electrode and cause a waste of energy.

3.2.3. Effect of pH value

pH value plays a significant role in the formation of hydroxyl radicals in EBCP, so it affects direct oxidation, indirect oxidation and microbial activity. The solution of acid or alkali has a great influence on the degradation of pollutants. The acidic solution easily controls oxygen evolution to enhance the overall current efficiency for both direct and indirect oxidation of organic pollutants on the anodes [98]. But with the extension of electrolysis time, hydrogen evolution reaction occurring at the cathode and a large number of H^+ are consumed to generate H_2, leading to an increase in pH and decrease in conductivity [99]. Meanwhile, alkaline solution helps to form carbonate and bicarbonate, which are well known scavengers for hydroxyl radical [100]. Simultaneously, pH can change the adsorption of the particle electrode. In practice, the adjustment of pH required more chemical reagents, operation costs, maintenance costs, and most microbes are suitable for survival in a neutral environment, so the working environment of EBCP cannot be too acidic or alkaline. Sun et al. [88] reported that the electrochemical treatment of chloramphenicol using Ti-Sn/c-Al_2O_3 particle electrodes with a three-dimensional reactor, high CAP degradation efficiency was obtained under acidic conditions. However, at pH = 6, the CAP degradation efficiency was as high as 71.3%. This value only decreased by 7.6% compared with that at pH = 2. Cao et al. [101] used the coupling of electro catalytic reduction and biodegradation process to treat nitrobenzene wastewater and studied the effect of pH on the removal rate of nitrobenzene. If there is no current in the reactor, nitrobenzene is completely removed by the microbial nitrobenzene reductase and pH value of 6 to 7 is the best. When current passes through the reaction system, in addition to participating in biological reactions, nitrobenzene is reduced near the cathode chamber by electro-catalytic reduction and the reaction rate is the largest at pH from 5.4 to 5.9. In conclusion, the presence of electric field changes the optimum pH range.

3.2.4. Effect of influent loading

Generally speaking, treatment efficiency decreased with the increasing of influent loading. When influent concentration is higher, the particle electrode will also partially lose adsorption and catalytic ability, and thus its service life becomes shorter. In other words, there is an optimum influent load in EBCP. Yang et al. [102] studied the effect of influent ammonia nitrogen load on denitrification in EBCP. It was observed that the total nitrogen removal was about 70%, when initial ammonia concentration was less than 70 mg/L. However, when initial ammonia nitrogen concentration is more than 120 mg/L, the ability to remove nitrogen is reduced because the metabolic capacity of microorganism has been restrained. In order to improve the removal capacity of ammonia nitrogen, greater current or longer processing time is required. Sreelatha et al. [103] evaluated the functional behavior of bio-electrochemical treatment system with increasing azo dye concentrations. Maximum dye removal was observed at 300 mg dye/l (75%) followed by 200 mg dye/l (65%), 100 mg dye/l (62%) and 500 mg dye/l (58%).

3.2.5. The other factors

The effects of electric field, hydraulic retention time, particle electrode dosage [20] and current action on the efficiency of an EBCP reactor are not well understood. The specific influence relationship and the rule between electric field and microbe are still unclear and need to be further studied in future work.

3.3. Environmental application

EBCP has been extensively studied for various wastewater treatment types, including inorganic wastewater, organic wastewater and some industrial wastewater. Table 2 sets out some typical cases of wastewater treatment by EBCP, which indicates that it offers hopes for future use.

3.3.1 Inorganic wastewater

3.3.1.1. Heavy metal wastewater

Mining operations discharging effluents, non-ferrous metal processing plants, electronic equipment factories and paint-coating workshops may produce higher concentrations of lead, copper, chromium, zinc, cadmium, arsenic, etc. These heavy metals cannot be degraded by microorganisms and can only be changed into different forms. They are difficult to eliminate by conventional physical-chemical treatment methods, such as chemical precipitation, membrane filtration, coagulation-flocculation, flotation, electrochemical, adsorption and ion exchange [114]. Although all heavy metal wastewater treatment techniques can be employed to remove heavy metals, they have their inherent advantages and limitations. The EBCP provides an effective method for the removal of heavy metals by the electrodeposition of cath-

Table 2
Typical case of wastewater treatment by EBCP

Wastewater	Contaminants	Reaction conditions	Residence time	Removal rate /%	References
	Acid Red	Initial mass concentration: 33.3 mg/L; Constant current: 15 mA;	6 h	Acid Red: 71.20	[30]
	Acid Red 18	Acid Red 18: 47.82–58.21 mg/L; Chroma: 466–728 mg/L; NH^{4+}–N: 7.22–14.57 mg/L; COD: 124.76–136.93 mg/L; Current density: 0.095 mA/cm^2;	Coupling hydrolysis: 12 h; Contact oxidation: 7.95 h;	Acid Red 18: 98.70; Chroma: 95.67; NH^{4+}–N: 53.15; COD: 90.06;	[104]
Printing and dyeing wastewater	Direct fast blue	Dyestuff mass concentration: 50 mg/L; Chroma: 130–150; NH^{4+}–N: 3 mg/L; COD: 70 mg/L ; Current density: 0. 096 mA/cm^2	Coupling hydrolysis: 12 h; Contact oxidation: 7.95 h;	Direct fast blue: 96.64; Chroma: 95.83; $NH4^{+}$–N: 51.43; COD: 87.10;	[76]
	Acid Red A	AR–A: 74 mg/L; COD: 88.6 mg/L; Methanol: 440 mg/L; Current: 300 mA;	7 h;	AR–A: 79.00; COD: 84.00;	[105]
	COD; Ammonia nitrogen; TP;	COD: 600–900 mg/L; NH^{4+}–N: 5–13 mg/L; TP: 10–20 mg/L; Current density: 3.636 mA/cm^2; Flow rate: 25 mL/min	Electrolysis: 28 min; Biodegradation: 100 min	COD: 80.90; NH^{4+}–N: 82.40; TP: 89.5;	[106]
Vanillin wastewater	Amino–dimethyl–aniline;	COD: 140 mg/L; Dimethyl–p–phenylenediamine: 10–4 mg/L; Current density: 50 mA/cm^2; Temperature: 24–26°C; pH = 6–7;	5 h	COD: 100;	[107]
Explosives wastewater	TNT	TNT: 0.32 mg/L; NH^{4+}–N: 4–116 mg/L; COD: 240–340 mg/L; pH = 7.0–7.5; Voltage: 24 V;	300 min	TNT: 77.90; NH^{4+}–N: 92.80; COD: 85.50;	[108]
Phenol wastewater	Pb2+; Phenol; Phenol;	Pb^{2+}: 50 mg/L; Phenol: 1500 mg/L Phenol: 200 mg/L; COD: 463.2 mg/L; Current density: 50 mA/cm^2; Temperature: 25°C;	3 h 120 min	Pb^{2+} : 99.00; Phenol: 99.90; Phenol: 100; COD: 94.00;	[109] [110]
	Phenol; Cr^{6+}; Zn^{2+};	Phenol: 2400 mg/L Cr^{6+}: 50 mg/L; Zn^{2+}: 125 mg/L; Electric field conditions: 3.0 V , 17.7 V/m, 1.98 A/m^2	57 h	Phenol: 100; Cr^{6+}: 99.90;	[111]
Wastewater containing phenanthrene	Phenanthrene; COD;	Phenanthrene: 20 mg/L; COD: 2000 mg/L; Sludge 3 g TSS/L; Temperature: (35 ± 1)°C; pH = 8.0; Current: 2 mA;	93 h	Phenanthrene: 99.40; COD: 91.50;	[112]
Urban sewage	COD; Nitrate nitrogen;	COD: 60–80 mg/L; NH^{4+}–N: 24–40 mg/L; Current density: 0.041 mA/cm^2	4 h	COD: 50.00; NH^{4+}–N: 82.04;	[113]

ode, the physical adsorption of biofilm and suspended substances. A similar phenomenon was observed in the removal of heavy metals. Liu et al. [109] utilized an electro-biofilm reactor to remove lead and phenol from wastewater. The concentration of lead and phenol decreased from 50 mg/L to 0.5 mg/L and from 1500 mg/L to 5 mg/ L, respectively. Wang et al. [111] developed a cathode biofilm fixed-bed reactor for the treatment of various heavy metal ions in wastewater. Depending on the cathodic electrodeposition effect, the maximum removal efficiency for Pb^{2+}, Zn^{2+} and Cd^{2+} was 99.8%, 98.6% and 88.3%, respectively. Watanabe et al. [115] observed that an electrode biofilm reactor is capable of denitrification while removing Cu^{2+} and Pb^{2+} ions, which indicates that EBCP provides an effective method for removal of heavy metal wastewater.

3.3.1.2. Nitrate wastewater

At present, the main point is autotrophic denitrification and simultaneous nitrification and denitrification (SND) in terms of the biofilm removing nitrogen. The nitrate removal in EBCP should be attributed to the autotrophic denitrification taking place thanks to hydrogen gas generated by a cathodic reaction, as the electron donor by autotrophic denitrifying bacteria and formation of anaerobic environment near the biofilm promote further denitrification [116]. A complete degradation of NO_3^- to N_2 gas by autotrophic denitrifying pathways was accomplished in four steps, evolving NO_2^- anions, nitric oxide (NO) and nitrous oxide (N_2O) gas as intermediates [117]. The subsequent reduction steps are carried out by denitrifying bacteria, which utilize nitrate and/or nitrite as elec-

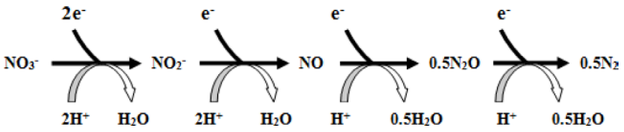

Fig. 2. Reaction process of NO_3^- conversion to N_2.

tron acceptors under anoxic conditions. The step-by-step biochemical reactions for the denitrification process are demonstrated in Fig. 2 [118–120].

SND refers to simultaneous nitrification and denitrification in a single reactor, in which a large number of tiny pure oxygen bubbles is generated by an inert anode and hydrogen is produced by a cathode. Subsequently, these gases are utilized by the nitrobacterium and denitrification bacteria which are embedded in the particle electrode and cathode surface [121]. The electrons produced through microbial respiration are accumulated on the surface of the anode and travel towards the cathode, where they are rescued by denitrifying microorganisms to degrade NO_3^- to molecular N_2 [117]. The total denitrification reaction equation is shown below:

$$NO_3^- + 5e^- + 6H^+ \rightarrow 0.5N_2 + 3H_2O \quad (3)$$

Sakakibara et al. [122] used the electrode biofilm method to treat NO_3^- in drinking water. It was observed that more than 95% NO_3^- was converted to N_2, but without N_2O or NO production. Tang et al. [123] successfully started up completely autotrophic nitrogen removal process by adjusting current load on the reactor to change dissolved oxygen and pH in cathode chamber, when the influent NH_4^+-N, pH and HRT (hydraulic retention time) were 35 mg/L,7.8 and 24 h, respectively. After 30 d of biofilm formation and 60d of adaptation, the removal rates of NH_4^+-N and TN (total nitrogen) reached 91% and 85%, respectively.

3.3.1.3. Phosphorus wastewater

The phosphorus degradation mechanism in EBCP is very complex, and in general, two different reinforcing ways are considered responsible for phosphorus degradation: (1) electric field improves the role of anaerobic releasing phosphorus and aerobic uptake of phosphorus; (2) the electric field also stimulates the activity of phosphorus accumulating organisms (PAOs). Tang et al. [124] studied the effect of electric field on biological phosphorus removal by contrast experiment, the average removal rate of total phosphorus (TP) in EBCP was 15% higher than in an ordinary biological reactor. In 2014, Wang et al. [125] studied the characteristics of denitrification and dephosphorization in a three-dimensional electrode biofilm reactor, using sponge iron or activated carbon composites as fillers. When electric current was 40 mA and HRT was 10 h, it was observed that the TP removal efficiency was more than 81%. Tao et al. [126] evaluated the effect of dissolved oxygen on nitrogen and phosphorus removal by two-chamber microbial fuel cell. It was observed that a great deal of TP was removed owing to chemical precipitation (about 80%) and microbial absorption (around 4–17%).

3.3.2 Organic wastewater

3.3.2.1. Dye wastewater

Dye wastewater in most cases is complex in structure and toxic, making it difficult to degrade by biological processes [127]. EBCP changes microbial cell membrane permeability under micro-current stimulation, and increases the contact area of the oxygen reductase with dye, making removal efficiency higher than in an independent electrochemical method, biological membrane method or both combined. Lang et al.[104] carried out experiments with bio-electrochemical technology for Acid Red 18 dye wastewater, testing the effect of different current densities on its treatment. When the current density was increased to 0.095 mA/cm^2, the removal efficiency of Acid Red 18, color and CODcr were 98.70%, 95.67% and 90.06%, respectively. Cao et al. [105] dispose of Acid Red A dye wastewater with a concentration of 74 mg/L through the EBCP. When the electric current was 300 mA and HRT was 7 h, the removal rate of CODcr was 84%, which was high 8% than without current.

3.3.2.2. Nitrobenzene wastewater

Nitrobenzene is an important chemical raw material for the production of aniline (AN), dyes, pharmaceuticals, and pesticides [128], but it is difficult for microorganisms to degrade. Many countries have listed it as a controlled pollutant, because it was considered to be harmful to animals. In EBCP, nitrobenzene was transformed into less-toxic and biodegradable aniline by electrocatalytic reduction [129], and then easily degraded by microorganisms so as to accomplish the purpose of efficiently removing nitrobenzene. Xu et al. [130] pretreated synthetic nitrobenzene wastewater by electrocatalysis using activated carbon as particle electrodes, so that the non-biodegradable nitrobenzene was converted into biodegradable aniline, and its biodegradability was enhanced. Cao et al. [101] used the electro-biofilm reactor to deal with nitrobenzene wastewater. With the current density increasing, the nitrobenzene removal efficiency and aniline production rate were also raised, and the nitrobenzene removal efficiency reached 95%, which is 46% higher than that without current. Zhang et al. [131] investigated the material's electro-reduction activity, with nitrogen-doped diamond electrode showing high performance for electrochemical reduction of nitrobenzene. Nitrobenzene removal efficiency and aniline formation efficiency were 96.5% and 88.4% under optimal conditions, respectively.

3.3.2.3. Refractory toxic organic wastewater

The refractory toxic organic wastewater (RTOW) is mainly composed of halogenated hydrocarbons, benzene ring and other organic compounds. Different research studies indicated that some of these molecules are recalcitrant and persistent in treated water because they are refractory to conventional biological treatment [132–134]. It is mainly their toxicity that completely or partially inhibits microbial metabolism (e.g. antibiotics, antiseptics, hydrophobic solvents, etc.). Their particular physical

properties, such as weak aqueous solubility or limited adsorption ability as well as complex molecular structure (e.g. carbamazepine, naproxen, etc.) make their chemical bonds difficult to break for all but few enzymes [135]. The RTOW degradation mechanism in EBCP is very complex, and in general two different ways are considered responsible for RTOW degradation: firstly, under electric current function, the reductive environment is formed near the cathode in EBCP, so that toxic substances such as halocarbons and benzene rings are reduced to bio-available intermediates, and then those intermediates are used as a carbon source or energy ,so as to achieve the purpose of efficient removal of organic wastewater. Xu et al. [136] studied the degradation process of chlorobenzene and dichlorobenzene by biofilm-electrode technology. Chlorobenzene and dichlorobenzene were preferentially dechlorinated and then degraded and open-looped. The formation of reduction environment by the cathode plays a key role in dechlorination process. Zhang et al. [137] and Zhang et al. [138] reported that hydrogen generated by the cathode can degrade dichlorophenol, dichloro-phenoxy acetic acid, trichloroacetic acid, and finally produce acetic acid. Secondly, the EBCP is an organic combination of redox with strong hydroxyl radical oxidation, producing non-biodegradable material ring capable of generating other substances, which contain hydroxyl and carboxyl compounds, (mineralized as H_2O and CO_2), and then used as a carbon source or energy by microorganisms. Chen et al. [139] treated 1.0 g/L L-leucine synthetic wastewater using activated carbon and ceramic particles as particle electrodes, respectively. Under optimum operating conditions, L-leucine and COD removal efficiency were 90% and 88%, respectively. At present, there are few studies on the second kind of speculation, so further experiments are necessary to provide conclusive proof.

3. Summary and perspective

EBCP is a process whereby electrochemical and microbial reactions are coupled in a single reactor capable of performing autotrophic denitrification, simultaneous nitrification and denitrification, and degradation of organic substances, to assist in the removal of biological phosphorus and heavy metal ions. EBCP heightens removal efficiency and reduces engineering costs. At the international level, the fundamental mechanism of EBCP is not properly understood. The factors contributing to pollutant removal are mainly focused on electric field strength, pH, temperature and electrode materials. However, with the underlying theory far from perfect, little is known about the mechanism of action of microorganisms with electric fields. EBCP exerts a stimulating effect on microbes through an electric field. The electric field has contributes to the growth of microorganisms, biological diversity and biological metabolism. The research on the biological effects of electric field is mainly focused on macroscopic biological effects, while the research on microscopic mechanism is still limited, hampering any further development of the EBCP. Therefore, there is an urgent need for studies of the microscopic mechanism from a molecular biology perspective, for analyzing the relationship between the electric field and microbial metabolism in order to enable the EBCP to be developed and utilized on a larger scale as well as to better understand the underlying theory. As an emerging technology, it still poses questions which need to be addressed:

(1) In terms of spatial structure, EBCP reactor enlargement and optimization are urgently needed. The ease of operation, superior removal efficiency and low investment costs are among the EBCP's key benefits that should be the focus of efforts to promote the technology to commercial use.
(2) As regards regulatory factors, an optimum control strategy should be formulated following a survey of the quantitative relationship between the key regulatory factors and pollutants.
(3) In discussing the mechanism of non-biological organic compound degradation , a variety of modern technologies should be utilized to reveal the synergic mechanism of the electrochemical and biological methods;
(4) The mechanism of microbial action should be discussed from the viewpoint of microbial ecology and molecular biology. The interaction mechanism between current and microorganisms should be clarified, and the stimulating effect of microbial population should be explained. Depending on the specific application, high-efficiency and high-activity functional microbes should be selected for the micro-electric field. The development of the corresponding high-current immobilized microbial treatment technology is required but the research in that area is still insufficient and needs to be intensified.

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