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Enhancing organic contaminant degradation through integrating advanced oxidation processes with microbial electrochemical systems

Kaichao Yang [a], Ibrahim M. Abu-Reesh [b], Zhen He [a,*]

[a] *Department of Energy, Environmental and Chemical Engineering, Washington University in St. Louis, St. Louis, MO 63130, USA*
[b] *Department of Chemical Engineering, Qatar University, Doha, Qatar*

A R T I C L E I N F O

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A B S T R A C T

Microbial electrochemical systems (MES) are studied to degrade organic contaminants with a lower energy demand, but degradation of recalcitrant compounds tends to be challenging. To enhance contaminant degradation in MES, advanced oxidation processes (AOPs) are synergistically linked to create cooperative processes such as bio-electro-Fenton (BEF) and enhanced bioanodes. BEF can achieve a high contaminant degradation efficiency with a low energy consumption due to the ability for energy recovery from the anodic organic wastes. Modifying a bioanode with catalytic oxidation materials, e.g., photocatalyst and MnO_2, will achieve organic removal via the cooperation of catalysis and biodegradation. This paper has provided a concise review on the integration of AOPs with MES and identified and discussed the challenges such as deeper understanding of the electron transfer mechanisms, development of low-cost membrane, and the synergetic effects between functional materials and bacteria that are important to develop AOP-MES treatment systems.

1. Introduction

Microbial electrochemical systems (MES) are an emerging technology for organic contaminant degradation. The anode/cathode in an MES can serve as effective electron acceptors/donors to promote the bioelectrochemical degradation process (Yang et al., 2022). However, the MES technology has limitations to degrade highly toxic and recalcitrant organic contaminants such as phenolic compounds and nitro aromatic compounds, because these contaminants can inhibit the activities of the functional (degradative and electroactive) bacteria and thus are difficult to remove in a benign biological process (Yang et al., 2021). To enhance the degradation of these organic contaminants in MES, advanced oxidation processes (AOPs) can be integrated. Electrochemically based AOPs such as electro-Fenton (EF) and electrochemical oxidation (EO) processes have been widely studied for their degradation performance through generating strong oxidants (e.g.,·OH and H_2O_2) continuously and on site. Those processes can achieve good organic degradation performance, but they are generally costly and energy-intensive (Li et al., 2018).

The cooperation between MES and AOPs creates a solution to overcome the drawbacks in a standalone process. Such a cooperation can be realized via either external connection or internal integration. MES can be externally connected to (electrochemically based) AOPs through an external circuit, where MES serves as a power source to provide electrons to the electrochemically based AOPs (Yuan et al., 2010; Zhu and Logan, 2013). Although the feasibility of this cooperation has been demonstrated, MES as a power source could be substituted by other power supplies like solar energy; therefore, this external connection is not unique and lacks a strong synergy between the two. The internal integration is to have AOPs inside an MES that allows the two processes to assist each other during contaminant degradation. Representative examples include the bio-electro-Fenton (BEF) process and the enhancement of bioanode. This paper aims to provide a concise review on those two types of internal integration between MES and AOPs and analyze the literature qualitatively and quantitatively. The challenges and opportunities of these integrations will be discussed.

2. Bio-electro-Fenton process

In a BEF process, easily biodegradable organics (e.g., municipal wastewater) are introduced into the anode chamber of an MES to provide electrons, which flow from the anode to the cathode via an external circuit and react with O_2 to produce H_2O_2 (Eq. 1) in the cathode chamber. To convert H_2O_2 into ·OH, strategies such as adding Fe^{2+},

* Corresponding author.
E-mail address: zhenhe@wustl.edu (Z. He).

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ultraviolet (UV) radiation, and O_3 can be applied (Eqs. 2–4) (Chen et al., 2021; Li et al., 2017; Zou et al., 2022). The generated ·OH in the cathode chamber is a powerful oxidant to degrade toxic and recalcitrant organic contaminants in a wastewater (which is different from the anode wastewater) (Fig. 1A). More details about the technology development, reactor design and affecting parameters of BEF can be found in several review papers (Li et al., 2020; Li et al., 2018; Zhao et al., 2021). We have summarized the studies using BEF to treat recalcitrant organic contaminants such as phenolic compounds, dyes, pharmaceuticals, and real wastewater like landfill leachate and oily wastewater (Table 1). Different contaminants have different environmentally relevant concentrations in actual wastewaters. Some wastewaters can have a high concentration of chemical oxygen demand (COD) for example landfill leachate at 2152 mg L^{-1} (Hassan et al., 2017), while the concentrations of pharmaceuticals are at ng-ug L^{-1} levels (Nadais et al., 2018; Santos et al., 2010). It should be noted that studies need to use environmentally relevant concentrations to make the obtained results useful to be extrapolated to the realistic wastewater. Concentrations higher than environmentally relevant concentrations could enlarge the removal rate due to the existence of more substrates, while a very low concentration may lead to more mass transfer limitations, resulting in the increase of energy consumption.

$$O_2 + 2H^+ + 2e^- \rightarrow H_2O_2 \quad (1)$$

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

$$H_2O_2 + h\nu \rightarrow 2\cdot OH \quad (3)$$

$$2H_2O_2 + 2O_3 \rightarrow H_2O + 3O_2 + HO_2\cdot + \cdot OH \quad (4)$$

The satisfactory removal of the target contaminant and COD/total organic carbon (TOC) from the polluted wastewater suggests that the BEF technology had a good degradation and mineralization ability (Table 1). A potential benefit of BEF treatment is its low energy consumption, largely related to the production of electrons from organic wastes in its anode. We have analyzed the available data from the previous studies shown in Table 1 and found that the mean and median energy consumption of typical BEF processes is 0.53 and 0.54 kWh kg^{-1}-target pollutant, respectively, an order of magnitude lower than those of conventional EF processes (32.61 and 30.15 kWh kg^{-1}-target pollutant, respectively) (Fig. 1B). The energy consumption data of EF were obtained from the studies degrading recalcitrant organic contaminants (e.g., phenolic compounds and azo dyes) at comparable concentration levels as mg-g L^{-1} (Liu et al., 2021; Nidheesh and Gandhimathi, 2012; Wang et al., 2021; Wang et al., 2023; Zhou et al., 2012). It should be noted that BEF exhibits lower removal rates of the organic pollutants than those in EF, likely due to its lower system current/voltage. For example, the removal rate of aniline in a BEF was 35 mg L^{-1} h^{-1} with an energy consumption of 0.728 kWh kg^{-1}-aniline (Li et al., 2017), while an EF could achieve 215 mg L^{-1} h^{-1} at an energy cost of 74 kWh kg^{-1}-aniline (Brillas and Casado, 2002). Despite having lower removal rates, the removal efficiencies of the target contaminant (mean and median values: 87.4 % and 95.5 %) and TOC/COD (mean and median values: 78.0 % and 81.0 %) in a BEF could still be close to those in an EF process (target contaminant removal: mean and median values as 87.5 % and 95.4 %; TOC/COD removal: mean and median values as 85.9 % and 91.0 %) (Fig. 1C). It should be noted that most studies of BEF were performed using a catholyte (e.g., Na_2SO_4 and phosphate buffer solution (PBS)) without other organic or inorganic components (Table 1). Although there are several studies treating real wastewater (e.g., oily wastewater and landfill leachate) using BEF, they mainly focused on the removal of the organic compounds, and the effect and fate of the inorganic constituents were ignored. The presence of the inorganic

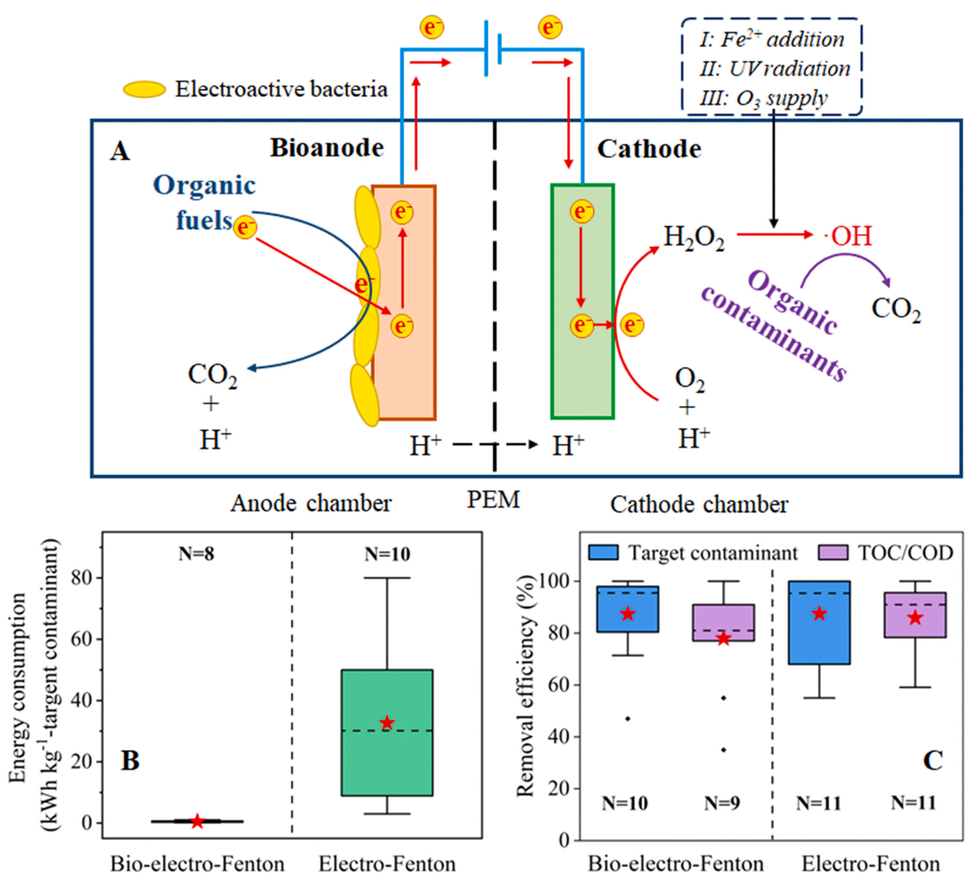

Fig. 1. Mechanisms of BEF process (A); the comparison of the energy consumption (B) and removal efficiency of target contaminant and TOC/COD (C) between the BEF and EF processes (red stars indicate mean values, black dashed lines indicate median values, and black dots indicate outliers).

Table 1
Bio-electro-Fenton for the degradation of organic contaminants.

Target pollutant	Concentration	Cathode electrode	Cathode solution	Target pollutant removal	Organic compounds removal	Reaction time	Reference
4-Nitrophenol	139 mg L^{-1}	Carbon felt	0.2 M Na_2SO_4 + 10 g scrap iron at pH 3[d]	100 %	85 % TOC	96 h	(Zhu and Ni, 2009)
4-Nitrophenol	35 mg L^{-1}	Carbon felt	1 M Na_2SO_4 + 112 mg limonite at pH 2[d]	96 %	NA	6 h	(Tao et al., 2013)
Triphenyltin chloride	35 mg L^{-1}	$Fe@Fe_2O_3$ graphite[a]	2 % NaCl at pH 3[d]	80.4 %	NA	100 h	(Yong et al., 2017)
Acid Orange 7	50 mg L^{-1}	Carbon felt	20 g L^{-1} Na_2SO_4 + 1 g $FeVO_4$ at pH 3[d]	89 %	81 % COD	60 h	(Luo et al., 2011)
Acid blue 113	100 mg L^{-1}	Graphite rod	0.1 M Na_2SO_4 + 30 mg L^{-1} $FeSO_4$ at pH 3[d]	71.4 %	55 % COD 29 % TOC	12 h	(Asghar et al., 2017)
Orange II	35 mg L^{-1}	γ-FeOOH/CNT[b]	100 mM phosphate buffer solution at pH 7	98 % at 12 h	91 % TOC at 43 h	43 h	(Feng et al., 2010)
Rhodamine B	15 mg L^{-1}	$Fe@Fe_2O_3$/NCF[c]	Synthetic solution at pH 3[d]	47 %	35 % TOC	12 h	(Zhuang et al., 2010)
Methylene blue	50 mg L^{-1}	Graphite	0.1 M Na_2SO_4 + 2 mM $FeSO_4$ at pH 3[d]	97 % at 8 h	99.6 % TOC at 16 h	16 h	(Zhang et al., 2015)
17β-estradiol 17α-ethynyl-estradiol	20 ug L^{-1}	$Fe@Fe_2O_3$/NCF[c]	0.1 M NaCl at pH 3[d]	81 % 56 %	NA	10 h	(Xu et al., 2013)
Ketoprofen Diclofenac Ibuprofen Naproxen	40 ug L^{-1}	Carbon felt	0.05 M Na_2SO_4 + 7.5 mM $FeSO_4$ at pH 2[d]	61 % 97 % 86 % 81 %	NA	5 h	(Nadais et al., 2018)
Oily wastewater	COD: 878 mg L^{-1}	Carbon felt	Real wastewater + $FeSO_4$ at pH 3 adjusted by H_2SO_4		77 % COD	183 h	(Wu et al., 2015)
Landfill leachate	COD: 1022 mg L^{-1}	Pyrrhotite-coated graphite sheet	Real landfill leachate at pH 3 adjusted by H_2SO_4		78 % COD	45 d	(Li et al., 2010)
Landfill leachate	COD: 2152 mg L^{-1}	Carbon felt	Real landfill leachate + $FeSO_4$ at pH 3 adjusted by H_2SO_4		41 % COD	NA	(Hassan et al., 2017)

[a] $Fe@Fe_2O_3$: Composite of Fe^0 and Fe_2O_3.
[b] CNT: Carbon nanotube.
[c] NCF: Non-catalyzed carbon felt.
[d] The pH was adjusted by spiking either H2SO4 in Na_2SO_4 catholyte, or HCl in NaCl catholyte. NA: Not available.

components not only could affect the organic degradation (e.g., NO_3^- may compete for the electron donors for H_2O_2 generation), but also generate more hazardous byproducts during BEF process (e.g., Cl^- and NH_4^+ may react with radicals and organics to form organic (e.g., haloacetonitriles) and inorganic (e.g., ClO_3^-, ClO_4^- and NO_2^-) byproducts) (Iskander et al., 2020).

Operating conditions (e.g., cathodic potential, current density, pH and cathode material) can be optimized to achieve a high contaminant degradation performance and low energy cost in BEF (Li et al., 2018). For example, cathodic potential, which is affected by electrode materials, can be controlled to improve the selectivity of H_2O_2 generation and it was found that the optimal potential was − 0.6 V (vs. SCE) for graphite cathode, − 0.8 V (vs. SCE) for graphite particle cathode, and − 1.0 V (vs. Ag/AgCl) for graphite carbon black hybrid cathode (Chen et al., 2014; Li et al., 2016; Sim et al., 2015). Enhancing current density under an optimized cathode potential would further improve the H_2O_2 production rate, thereby increasing contaminant degradation. Computational tools can help identify the most critical parameters affecting the BEF performance, as well as guide the system design and parameter optimization (Gadkari et al., 2018). Furthermore, several Fe-functionalized carbonaceous cathodes such as $Fe@Fe_2O_3$/graphite ($Fe@Fe_2O_3$ refers to the composite of Fe^0 and Fe_2O_3), γ-FeOOH/carbon nanotube, and $Fe@Fe_2O_3$/non-catalyzed carbon felt were successfully designed and served as a solid Fe catalyst source to avoid the need for UV/O_3 addition, resulting in a lower energy cost (Feng et al., 2010; Yong et al., 2017; Zhuang et al., 2010) (Table 1).

Although efforts have been invested to improve the rate of H_2O_2 generation and the production of ·OH in BEF, several critical challenges remain to address. First, BEF reactors require expensive membrane (e.g., proton exchange membrane) to separate the cathode and anode chambers to maintain the bioanode activity. Because the cathodic environments (i.e., H_2O_2 accumulation, acidic pH condition, and Fe^{2+}, UV or O_3 addition) for a Fenton process are unsuitable for the growth of anodic functional bacteria, it is not feasible to eliminate membrane at this moment. Low cost and durable membranes can help reduce the cost of BEF technology and rapid progress in material science will accelerate membrane development. Second, when treating highly toxic and recalcitrant organic contaminants (e.g., industrial wastewater) in the cathode of an BEF, easily biodegradable organic compounds (e.g., municipal wastewater) will be needed as an electron source in the anode chamber. That requires the co-location of sources that generate different types of wastewaters or infrastructure to transport those wastewaters to the treatment site. That is possible in some municipal wastewater treatment plants that receive industrial effluents, but such a requirement will limit the application scope of BEF or add more cost to its operation. Third, the generation/accumulation of byproducts during the BEF process needs to be further understood. Fenton's reaction is nonselective and can produce oxidation byproducts, many of which are highly toxic or carcinogenic (Iskander et al., 2020). Further disposal of the cathode effluent, for example through the anode for bioelectrochemical treatment, warrants investigation (Yang et al., 2022). Finally, Fenton's reaction could result in the accumulation of sludge that needs to be properly managed.

3. Enhancement of the MES bioanode

To make the operation simpler (in comparison to BEF), a desired approach is to treat the recalcitrant compounds in the anode, where they act as an electron source. This can be realized by enhancing the degradation ability of a bioanode through coating catalytic oxidation materials like photocatalysts or catalysts. Because such an enhanced process is typically driven by photo/electric energy, no chemical addition will be needed, thereby reducing the risk of secondary pollution compared to BEF.

Photocatalysts can be coated onto the bioanode of an MES to generate reactive oxygen species (ROSs) (mainly ·OH) that assist the biodegradation process for the removal of organic contaminants. The noteworthy problem is that the ROSs generated by the photocatalysts may also damage the functional bacteria due to their direct contact in the bioanode. To address that problem, an intimately coupled photo-biocatalysis (ICPB)-anode was designed (Zhou et al., 2017). In a system containing an ICPB-anode, the functional bacteria were located away from where the ROSs were generated, yet close enough to quickly biodegrade the intermediate organics produced from the photocatalysis and thus generate bio-electrons (Fig. 2A). For example, photocatalytic oxidation and biodegradation could cooperate well on an ICPB-anode to achieve a sequential oxidative ring-cleavage and mineralization of 4-CP (Fig. 2B). Our analysis of the previous studies using ICPB-anodes shows a significantly improved degradation efficiency compared with that using conventional bioanodes ($p < 0.05$) (Ding et al., 2018; Long et al., 2020; Shi et al., 2022; Zhou et al., 2018; Zhou et al., 2017). The mean and median values of the target organic pollutant removal efficiency by the ICPB-anodes were 81.9 % and 85.3 %, respectively, double those with the conventional bioanodes (42.5 % and 40.0 %, respectively) (Yang et al., 2022; Yang et al., 2020) (Fig. 2C). Although the removal efficiencies of TOC/COD by the ICPB-anodes (mean and median values as 51.0 % and 30.1 %) were also obviously enhanced compared to the conventional bioanodes (mean and median values as 28.6 % and 22.0 %), the mineralization ability of the ICPB-anodes still needs further improvement (Fig. 2C). In addition, it should be noted that, up to now, all the studies about ICPB-anodes used external lamps for illumination, which leads to another energy consumption source, photoenergy. The consumption of photoenergy needs to be further analyzed, unless natural sunlight is used. The development of innovative anode material may facilitate the use of natural sunlight to further reduce the energy consumption.

In addition to photocatalysts, catalytic oxidation materials like MnO_2 were also used to modify bioanodes to improve the recalcitrant organic oxidation (Chen et al., 2017a; Chen et al., 2017b; Yang et al., 2020). Unlike photocatalysts that utilize the ROSs for degradation, the oxidation with MnO_2 ascribes to the high redox potential of φ (MnO_2/Mn^{2+}) (1.23 V), which is high enough to oxidize a variety of aromatic organic compounds, such as phenolic compounds, aromatic amines, and aromatic thiols (Grebel et al., 2016; Stone, 1987). For example, phenol wastewater was directly used as a fuel in the MES bioanode modified with MnO_2 that achieved phenol removal and current generation via the cooperation of MnO_2 oxidation and biodegradation processes (Chen et al., 2017a; Chen et al., 2017b). The phenol degradation efficiency and current generation were increased by about 73.3 % and 104.3 %, respectively, compared with those of a conventional bioanode (Chen et al., 2017b). Moreover, MnO_2 has the ability to store electrons according to Eq. 5 (Li et al., 2014). When electron acceptors for the MES bioanode become unavailable, the electrons stored in the unstable $MnOOM_xH_y$ could be released (Eq. 5) and transferred to the bioanode to maintain the system current, thereby promoting substrate degradation in turn.

$$MnO_2 + xM^+ + yH^+ + (x+y)e^- \Leftrightarrow MnOOM_xH_y \ (M^+ = Na^+, K^+, \text{or et al.}) \quad (5)$$

Although the enhanced bioanode exhibits the ability to improve the degradation performance of toxic and recalcitrant organic contaminants in MES, this approach is relatively new and has some key questions to answer. First, the specific electron transfer mechanism between catalytic materials and functional bacteria is yet to be understood. It was reported that the electroactive species *Geobacter* had the ability to transfer the photo-generated electrons with an ICPB-anode, which helped improve the columbic efficiency and current generation of the MES (Zhou et al., 2018). By understanding the electron transfer mechanism and synergic effects between material catalysis and bacterial degradation processes, we may be able to optimize the cooperation to further improve the bioanode performance. Second, the stability of the enhanced bioanode needs to be demonstrated. The long-term effects of the coated catalytic materials on the microbial community and MES operation have not been well studied. Modeling could be a powerful tool to assist with the in-depth study of the enhanced bioanode. In recent years, modeling has attracted great attention due to its ability to help understand and optimize the MES bioanode system. Through the utilization of computational tools, we could not only gain a detailed insight into the internal processes and mechanisms (e.g., mass transfer/diffusion process, electron transfer mechanisms and redox reaction kinetics, and biofilm growth and bacterial transformation processes) (Liu et al., 2022; Ortiz-Martínez et al., 2015; Yahya et al., 2015), but also predict the performance and behavior of the bioanode in different condition and perform parameter optimization (Xia et al., 2018). Because MES bioanode is a highly complex and multivariable system that contains many biological, physical-chemical and electro-chemical processes, the modeling requires interdisciplinary knowledge and multi-disciplinary approaches. Finally, experiments need to be conducted under environmentally relevant conditions so that the results can have more environmental applications. In addition, almost all the existing studies of the enhanced bioanodes were conducted in dual-chamber reactors. Single-chamber MES can be of interest due to their lower cost and simpler structure for scaling up. However, without membrane separation, the possible interactions between the two electrode reactions and the effect of cathodic reactions on the contaminant degradation become more complex.

Both integration processes, BEF and enhanced bioanode, could achieve the effective removal of the recalcitrant organic contaminants.

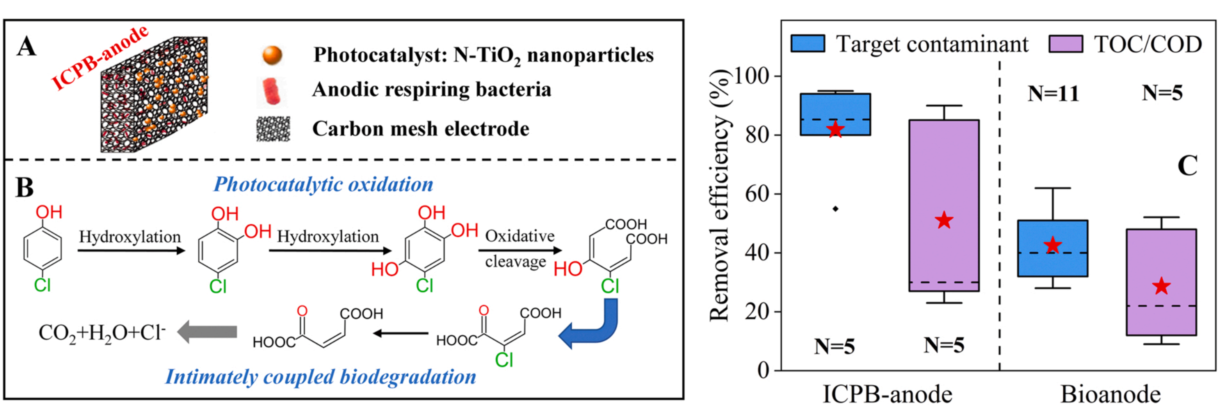

Fig. 2. Schematical illustration of the ICPB-anode (A); the degradation pathway of 4-CP in the ICPB-anode (B); and the comparison of the removal efficiency of target contaminant and TOC/COD between ICPB-anodes and conventional bioanodes (red stars indicate mean values, black dashed lines indicate median values, and black dots indicate outliers) (C).
Fig. 2A and B Reproduced from Zhou et al. (2017).

Compared to the enhanced bioanode (mean and median TOC/COD removal values as 51.0 % and 30.1 %), BEF had a relatively higher mineralization performance (mean and median TOC/COD removal values as 78.0 % and 81.0 %), likely due to the generation of more radicals from the Fenton process (Liu et al., 2021). However, BEF is a more complicated process that requires expensive membranes and different water sources. The enhanced bioanode, on the other hand, is more facile to be applied in membrane-free reactors and achieve energy recovery from treating wastewater. Moreover, the integration processes of MES and AOPs may significantly reduce the energy consumption compared to the electrochemically based AOPs (e.g., EF), while achieving a comparable removal and mineralization efficiency of the recalcitrant contaminants.

4. Summary and outlook

Integrating AOPs with MES has a great potential for enhancing the organic contaminant degradation. Internal integration can create a stronger synergy between the two processes. BEF process could have a low energy consumption, compared to EF, but have challenges like the use of separation membranes and requirement of different-type wastewaters. The enhancement of MES bioanode is accomplished by modifying the bioanode with catalytic oxidation materials. In this way, catalysis and biodegradation can be integrated to achieve joint efforts for organic contaminant removal. Further improvements of the enhanced bioanode need a better understanding of the electron transfer mechanisms and the cooperative effect between the catalytic oxidation and biodegradation processes. The cost of such technologies could be one of the decisive factors for their commercialization. Both capital and operation costs, which were rarely reported previously, should be accurately provided and analyzed in future studies. The development of low cost, durable and effective membrane and electrodes are essential to reduce the overall cost. The use of alternative energy such as renewable energy can make this approach more attractive. Additional benefits of the technology should be further explored, for example ·OH as a strong oxidant can also help with disinfection.

Declaration of Competing Interest

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

Data Availability

Data will be made available on request.

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