State-of-the-art review and bibliometric analysis on electro-Fenton process

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Abstract
The electro-Fenton (EF) process was first proposed in 1996 and, since then, considerable development has been achieved for its application in wastewater treatment, especially at lab and pilot scale. After more than 25 years, the high efficiency, versatility and environmental compatibility of EF process has been demonstrated. In this review, bibliometrics has been adopted as a tool that allows quantifying the development of EF as well as introducing some useful correlations. As a result, information is summarized in a more visual manner that can be easily analyzed and interpreted as compared to conventional reviewing. During the recent decades under review, 83 countries have contributed to the dramatic growth of EF publications, with China, Spain and France leading the publication output. The top 12 most cited articles, along with the top 32 most productive authors in the EF field, have been screened. Four stages have been identified as main descriptors of the development of EF throughout these years, being each stage characterized by relevant breakthroughs. To conclude, a general cognitive model for the EF process is proposed, including atomic, microscopic and macroscopic views, and future perspectives are discussed.

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Graphical abstract

Hot topics for future research revealed by the bibliometric analysis

Keywords Bibliometric analysis · Cathode · Fenton catalyst · Electro-Fenton · Hydrogen peroxide · Oxygen reduction reaction · Wastewater treatment

1 Introduction

The electro-Fenton (EF) process has been deemed a promising technology for water decontamination in recent years, owing to relevant merits like high efficiency, versatility and eco-friendliness [1]. In the classical EF process (Fig. 1), \( \text{H}_2\text{O}_2 \) is formed via cathodic two-electron oxygen reduction reaction (2e\(^{-}\)-ORR) (Eq. 1); for this, gaseous oxygen (O\(_2\)\(_{\text{gas}}\)) must be dissolved into the supporting electrolyte (O\(_2\)\(_{\text{bulk}}\)), then it is transported from the bulk solution to the diffusion layer, the double layer and, finally, it reaches the active sites at the cathode surface, where the ORR occurs. After this, the produced \( \text{H}_2\text{O}_2 \) diffuses away from the porous cathodic structure, undergoing catalytic decomposition upon Fenton’s reaction with soluble iron ions (Eq. 2), yielding the powerful oxidant hydroxyl radical (\( E_0(\cdot\text{OH}|\text{H}_2\text{O}) = 2.8 \text{ V vs. NHE} \)) through the Haber–Weiss mechanism [2]. Unavoidably, \( \text{H}_2\text{O}_2 \) is partially reduced to \( \text{H}_2\text{O} \) due to its confinement within the cathode pores. It has been proven that a vast majority of organic pollutants can be mineralized into inorganic ions and \( \text{CO}_2 \) under the action of \( \cdot\text{OH} \), thus being highly adaptable for the treatment of wastewater from different origin [3]. In general, the efficiency of EF is much higher than that of chemical Fenton process because Fe\(^{3+}\) can be continuously reduced to Fe\(^{2+}\) at the cathode surface (Eq. 3), thus minimizing the need for external addition of Fe\(^{2+}\) catalyst [4]. Therefore, both the 2e\(^{-}\)-ORR and the cathodic iron reduction are affected by electron transfer rate at the cathode/electrolyte interface and mass transport, as illustrated in Fig. 1. The \( \text{H}_2 \) evolution (Eq. 4) and the four-electron
oxygen reduction reaction (4e\(^-\)-ORR, Eq. 5) are typically the competing cathodic reactions that cause a decrease in current efficiency.

The efficiency of EF is mainly dependent on the cathodic H\(_2\)O\(_2\) production. Consequently, in recent years, a growing body of reviews has focused on this reaction, trying to address concerns related to low reactivity/selectivity and limited oxygen mass transport [5–7]. Reviews on other aspects of EF such as the effect of surface modification of carbonaceous cathodes [8, 9], reaction mechanisms, influence of key operation parameters, reactor design [10], coupling with high oxidation power anodes like boron-doped diamond (BDD) thin films [11], development of heterogeneous catalysts [12] or treatment of mixtures of pollutants, from sanitary landfill leachates [13] to arsenic [14], have been previously published. However, those reviews summarize the recent development of the EF process using the conventional reviewing method. Unlike the common summarizing approaches, bibliometric analysis with data visualization and specialized software (VOSviewer, Arnetminer, PaperLens, CiteSpace, etc.) allows presenting the literature in a more straightforward and meaningful way [15]. For the bibliometric analysis, statistical data analysis is performed on the basis of the year, country or institution, whereas a bibliometric network analysis is carried out from keyword occurrence, co-authorship and country collaboration. In this way, researchers can quickly find the outputs and cooperation of target authors or publications they intend to cite and, additionally, they obtain information to figure out hot topics for future research. Therefore, this bibliometric analysis offers a new approach to visually quantify the information on the development of EF over the past two decades. The review is timely because of the lack of such type of summary about EF, being possible to identify four stages of EF development based on a mathematical tool instead of the manual or mechanical methodology [16].

The bibliometric analysis, focused on the database from the core collection on the Web of Science aiming to ensure the integrity and academic quality, has been carried out with VOSviewer software [17]. Top keywords were analyzed to discern the progress in the country-wise distribution during the past 20 years. In addition, the most cited publications have been monitored and a general cognitive model for the EF process have been proposed for the first time.

\[
\begin{align*}
\text{Eq. (1)} & : O_2 + 2e^- + 2H^+ \rightarrow H_2O_2, \\
\text{Eq. (2)} & : H_2O_2 + Fe^{2+} \rightarrow Fe^{3+} + \cdot OH + OH^- \\
& \quad \quad \quad \quad \quad (\text{Absolute rate constant } (k_1) \approx 70 \text{ M}^{-1} \text{s}^{-1}), \\
\text{Eq. (3)} & : Fe^{3+} + e^- \rightarrow Fe^{2+} (E^0 = 0.77 \text{ V vs. NHE}), \\
\text{Eq. (4)} & : 2H^+ + 2e^- \rightarrow H_2, \\
\text{Eq. (5)} & : O_2 + 4e^- + 4H^+ \rightarrow 2H_2O.
\end{align*}
\]

2 Developments in the EF process in the past two decades

2.1 Trend in publication of articles in scientific journals

Up to 3012 research works from 83 countries have been published including “electro-Fenton” as keyword within the
The evolution of the number of publications on EF process is depicted in Fig. 2a, evidencing an exponential increase. In the first decade (2002–2012), only 355 publications were released, just accounting for 11.7% of the total number of works in the last 20 years. The year 2013 was especially significant because the yearly number of publications has been over 100 since then. The current average annual growth rate is 18.3%. Driven by the growth of green electrical power, ease of operation and relatively simple scale application upon cell stacking, the EF process has gained attention among the advanced oxidation processes (AOPs). To evaluate the quality of publications, the top 13 most productive countries along with their citation number per publication are presented in Fig. 2b. There is no doubt that the People’s Republic of China has been the most productive country, followed by Spain and France, during the past 20 years. However, when comparing the impact of each publication in terms of citations, Italy turns out to be the top one, followed by Spain and France, thus accounting for the high and worldwide relevance of scholars from these three countries. The collaborative networks between the top 15 most productive countries are highlighted in Fig. 2c. Among them, circles are representative of countries, in which a larger circle denotes a higher number of publications in that country. In addition, curves stand for more repeated relationships between two countries and thicker ones suggest stronger collaborations between them. As deduced from Fig. 2c, the strongest collaboration is established between China and USA, followed by a strong collaboration between China and Mexico. It can

**Fig. 2**  
(a) The number of publications between 2002 and 2022.  
(b) The first 13 most productive countries during the past 20 years along with the number of citations per publication.  
(c) Collaboration network between the top 15 most productive countries

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be noticed that Spanish research groups specialized in EF, with preponderance of the group from Prof. Brillas and Sirés, have a solid cooperation with France, Brazil, Mexico and Italy.

2.2 Four stages of the EF process development

Based on the total number of 3012 research works exported from the Web of Science Core Collection (period 2002–2012), the progress of the EF process can be divided into four stages, as depicted in Fig. 3. The main outcome in each of these four periods is highlighted as well.

2.2.1 The first stage: treatment of several pollutants

The first stage dates back to 1996 when the term “electro-Fenton” was first used by Prof. Brillas [18]. After that, Prof. Brillas’ and Prof. Oturan’s groups dominantly contributed to the work in the EF process, concentrating on the application of EF to the degradation and mineralization of various pollutants. These included herbicides (imazapyr [19], 4 chlorophenol [20]) and synthetic dyes (azo dyes [21], bromopyrogallol red [22], acid chrome blue K [23], etc.), being herbicides the main representative contaminants under study. Inspired by the innovative works from the both professors, EF process has been used to treat diverse wastewater afterwards. The feasibility and high effectiveness have been confirmed for leachate [24], persistent organic pollutants (POPs) [25, 26], green table olive processing wastewater [27], etc. During the first stage (1996–2006) of EF development, the mainstream work was related to the optimization of the key parameters that determined the performance for the treatment of organic pollutants, thereby being compared to other electrochemical advanced oxidation processes (EAOPs) for organic matter degradation, including anodic oxidation, photoelectro-Fenton and electrocoagulation. Among these, photoelectro-Fenton showed a much higher efficiency due to photoreduction of [Fe(OH)]2+ and photodecarboxylation of Fe(III)-carboxylate complexes [28]. The most influential parameters under optimization were the applied current density or electrode potential, solution pH, nature of supporting electrolytes, type and concentration of Fenton catalyst, temperature, oxygen or air flow rate and electrolysis time [1]. After that, the possible degradation pathways based on the detected byproducts using conventional and emerging technologies, like high-performance liquid chromatography (HPLC) and gas chromatography-mass spectrometry (GC/MS), were elucidated. In a word, literature about the EF process in the first stage was mainly focused on the engineering perspective, while the fundamental mechanisms the involving 2e−-ORR and Fe2+ regeneration reduction were much less investigated.

2.2.2 The second stage: three emerging areas

The second stage of EF development was followed the first stage during 2006–2007. Three research subfields have emerged since scholars started to devise strategies to enhance the performance of EF after 10 years development at the first stage, aside from extending its application to other types of wastewater. The three emerging areas (see Fig. 3) are described below:

(I) Cathodic H2O2 electrogeneration started to arouse researchers’ attention from a materials science standpoint, based on a structure-determined performance principle [29]. Hence, new cathodes with desirable structures instead of commercial ones (e.g., carbon-PTFE) were fabricated to boost the H2O2 formation, such as a novel gas-diffusion electrodes made from graphite powder and PTFE.
dispersion [30, 31], carbon nanotubes (CNTs) and core–shell iron-based particles like Fe@Fe₂O₃ nanowires-coated oxygen-diffusion electrodes [32].

(II) Mineral iron catalysts like goethite, alpha-FeOOH, magnetite (Fe₃O₄) and wustite (FeO) were proposed as an initial heterogeneous catalyst prototype to alleviate acidic pH limitation [33].

(III) Novel reactor designs were put forward. For example, a flow electrochemical reactor with reticulated vitreous carbon cathode [34], or a system in which the H⁺ and OH⁻ released at the electrodes were used to alleviate the pH limitation, thus avoiding the addition of chemicals [35].

2.2.3 The third stage: mechanism investigations

The third stage of EF development was between 2007 and 2018, as reflected by the co-occurrence network of the most frequently employed keywords (Fig. 4). The keywords co-occurrence network is a visualization tool to present the focus of a specific area [36]. In 2018, the keyword “hydrogen peroxide” was observed 16 times, “oxygen reduction reaction (ORR)” was employed 12 times and “wastewater treatment” appeared 20 times, in agreement with the large circles in green and yellow color in Fig. 4. However, the keyword “oxygen reduction reaction” failed to be observed in the co-occurrence network in 2014–2017, as indicated in Fig. SI 1–4. It means the oxygen reduction reaction for H₂O₂ production in the context of EF process has not attracted great attention until 2018.

Before 2018, the measured H₂O₂ concentration in EF publications was the accumulated one, according to Eq. (6) [37], which subtracts the decomposed H₂O₂ ([H₂O₂]decomposed) from the generated one ([H₂O₂]generated). Therefore, a decrease of the latter and a reduction of the former should boost the H₂O₂ accumulation. Regarding the methods to alleviate the H₂O₂ decomposition, pulsed electrolysis and unconventional supporting electrolyte
have been previously reported [38, 39]. Pulsed electrolysis replacing constant current/potential mode may hinder the decomposition thanks to the enhancement of products/reagents toward/from the cathode vicinity during the off-time period and thus, reduce their further reduction on the porous surface [40, 41]. In addition, unconventional pyrophosphate supporting electrolyte has been observed to decrease the decomposition rate of H2O2 [38].

The rate of [H2O2]generated via 2e−-ORR can be expressed as Eq. (7) [42], where \( k_{\text{het}} \) is the surface rate constant for electron transfer and \([O_2]\) is the local oxygen concentration at the cathode vicinity ready for 2e−-ORR. The rate constant is determined by the chemical catalytic effect on the cathode. As a consequence, strategies to improve interfacial electron transfer along with local O2 concentration will definitely boost the rate of 2e−-ORR for H2O2 accumulation. For example, a gas-diffusion electrode [43], a floating electrode [44], a pressurized jet aerator [45] and a microbubble-assisted cathode [2] were previously proposed to enhance the H2O2 accumulation mainly by increasing the local oxygen concentration. But only local oxygen concentration enhancement is not enough for H2O2 boosting since both surface charge transfer and oxygen concentration determine the occurrence of the ORR, as indicated in Eq. (7) [39]. To increase the activity and selectivity towards the 2e−-ORR, cathodes or electrocatalysts must endow suitable binding energy of *OOH intermediate to undergo a single-electron reduction (Eq. (8)), in which end-on adsorption of O2 on active sites is desirable for the first electron transfer step to keep the O–O bond in *OOH, as deduced from Sabiner’s principle for 2e− −ORR [46]. In addition, further dissociation steps of O–O bond in *OOH to tune the desorption of *OOH for H2O2 production rather than dissociation of O–O bonding with H2O production should be guaranteed as well (Eqs. 8–11). Hence, a series of publication on cathode design with diverse structures have been booming, reporting the introduction of defects [47], N-functional groups [48–50], graphene [43, 51, 52] and CoS2 nanoparticles [53]. Moreover, a series of focused reviews on cathodic H2O2 generation via 2e−-ORR has been published [6, 7, 54].

\[
[H_2O_2]_{\text{accumulated}} = [H_2O_2]_{\text{generated}} - [H_2O_2]_{\text{decomposed}},
\]

\[
k_{2e-\text{ORR}} = k_{\text{het}} \times [O_2],
\]

\[
*O_2 + H^+ + e^- \rightarrow *OOH,
\]

\[
*OOH + H^+ + e^- \rightarrow H_2O_2 + *,
\]

\[
*OOH + H^+ + e^- \rightarrow O + H_2O,
\]

Apart from the ORR-to-H2O2 green cluster shown in Fig. 4, the keyword “heterogeneous catalysts” increased its occurrence in publications since 2017–2018, reaching 44 publications in 2018 and 80 in 2022, as can be observed in the yellow cluster that appears in the co-occurrence network of the top keywords. Heterogeneous catalysts in the EF process have been booming at this stage to solve the limitations that are inherent to the conventional EF process, such as acidic pH restriction and slow catalyst recycling [55]. Based on their physical nature, heterogeneous catalysts for EF can be divided into the following four categories (Fig. 5), as listed below, whereas the preparation methods and underlying mechanisms have been systematically discussed in the literature [12, 55]:

(I) Iron minerals: pyrite (FeS2) [56], magnetite [57], hematite [58], goethite (α-FeOOH) [59], wüstite [60], and lepidocrocite [61],

(II) Zero-valent iron (ZVI) [62, 63], MOF-based ZVI [64]; iron foam [65, 66],

(III) Iron supported on synthetic structures like: organic polymers (alginate beads [67], chitosan [68]), inorganic substrates (nickel foam [69], graphene oxide [70], activated carbon [71], N-doped hierarchically...
porous carbon [72], hollow sea-urchin-shaped carbon [73]),

(IV) Iron supported on waste (rice straw, coal fly ash [74], acid mine drainage [75], industrial pyrite waste slag [76], zeolite [77]) and iron-rich soil (sepiolite [78], bentonite [79] and kaolin [80]).

2.2.4 The fourth stage: integral assessment

After more than 20 years of development, EF has gained growing attention both from fundamental and application point of view. As a consequence, we have devised a general cognitive model for the EF process based on the distinct stages of development. Three different scales are distinguished (Fig. 6):

(I) At the atomic scale, the regulation of the electronic effects is considered. This regards the creation of active catalytic sites at the atomic level both for the $2e^-$-ORR (Eq. 1) and iron reduction (Eq. 2). For this, the composition and morphology of the cathodes must be controlled. Regarding the $2e^-$-ORR, a growing body of literature has focused on the enhancement of the H$_2$O$_2$ accumulation by developing cathodes with different active sites [6, 7], introducing functional groups (O-, N-, F-, S-, P- and B-), defects, single atoms, etc. [81–86] (Fig. 7a).

Aiming to alleviate the slow cathodic iron reduction in the EF process, the iron-reducing bacteria Shewanella [87], dual cathode systems and pulsed current [37] have been recently explored. This is a serious issue, but not much attention has been paid so far because the cathodic Fe$^{3+}$ reduction in EF has been assumed as a comparable phenomenon to the homogenous iron reduction in non-electrochemical Fenton-based processes. Nonetheless, cathodic iron reduction has its particularities since electron kinetics, diffusion, and hydrodynamics determine the extent of Fe$^{2+}$ regeneration. Another emerging area in EF is the usage of density functional theory calculations (DFT) coupled with other quantum-based modeling strategies (like Gaussian, etc.), to bring insights into the electronic effects at the cathode and during organic degradation, at the atomic level [88–90]. The first DFT publication used in the EF process was reported by Mu’s group (Fig. 7b), revealing the charge redistribution using Pd-Fe$_3$O$_4$ catalyst [91].

(II) The microscopic scale involves the cathode/ electrolyte interface, where reactions occur in the so-called microenvironment. Hence, the main purpose of controlling the microenvironment is to coordinate the reagents (H$^+$/OH$^-$/O$_2$/Fe$^{2+}$) during the $2e^-$-ORR and cathodic iron reduction, coupled with the effects of cations (Na$^+$, K$^+$, etc.) and water dipole in the double layer. Both mass trans-
port and activity/selectivity may be tuned in this microenvironment. In this regards, superaerophilic electrodes with coarse nanoarray structures are of interest because they allow trapping oxygen inside textured surfaces [92]. The trapped oxygen, even in the submerged supporting electrolyte, is able to form the solid/liquid/gas tri-phase interface and then favors interfacial oxygen transport and electron transfer processes. As indicated in Fig. 7c, the degree of water intruded into the pores gives rise to the underwater Cassie (UC), underwater Wenzel (UW) and the underwater Wenzel-Cassie (UWC) wetting states, in which UWC accounts for the largest tri-phase interface. Results show that superaerophilic electrodes could change from the UWC to the UW state during electrolysis and thus, \( \text{H}_2\text{O}_2 \) production is affected.

(III) The macroscopic scale deals with the optimization of EF process for its large-scale application. Energy consumption is a crucial parameter when taking into consideration the scale-up step. For its minimization, several reactor configurations have been designed and sometimes modeled, such as the jet aerator reactor [93], a reaction with cation exchange resin that supplied Fe(II) [94], the rotating cylinder electrode (RCE) reactor [95] and a vertical-flow through reactor [96, 97], among others. Recent pilot-scale works have been reviewed by Casado [98]. Aside from reactor optimization and design, the bioelectro-Fenton system is another feasible way to lower the energy needs, since anodic electrogenic microorganisms produce electrical power from organics degradation to drive the Fenton’s reaction in the cathode chamber. The recent development of this promising process has been reviewed elsewhere [99, 100]. The usage of external fields like renewable sunlight [28] and magnetic forces [101], coupled with strategies to decrease the overpotential for anodic oxygen reduction reaction, is also under development. The lifespan of the cathode is crucial as well, but the investigation of its corrosion and fouling is still in its infancy [102]. Regarding the application of EF process to the treatment of emerging organic pollutants (antibiotics, personal care products), it must be noticed that this method has been also adapted to disinfection, especially after the Covid-19 pandemic (Fig. 7d). For example, EF has been used in inactivation of helminth eggs, Escherichia coli [103] and municipal secondary effluent [104].
2.3 Monitoring the references with strong citation bursts

Strong citation bursts for certain publications could be an indicator of a relevant discovery, thus being crucial for progress in a particular area [107]. Hence, citation bursts were analyzed based on the 3012 documents exported from the Web of Science. Here, only the top 12 articles that have received more than 500 citations are summarized in Table 1. Note that all of them correspond to review papers. In 2006, Prof. Pignatello published the review entitled “Advanced oxidation processes for organic contaminant destruction based on the Fenton reaction and related chemistry” in Critical Reviews in Environmental Science and Technology, which has been cited 2628 times. However, the first and most remarkable focused review paper on the EF process was published in 2009 by Profs. Brillas, Sirés and Oturan in Chemical Reviews [1]. Indeed, the “electron-Fenton” concept was first devised by Profs. Brillas and Oturan [18, 28], pioneers in the field. After this highly significant review, both groups have developed remarkable work in EF, laying the foundations of the process. In addition, other two reviews published by Prof. Sirés [108, 109] were highly cited as well. It can be seen in Fig. 2c that Spain and France have the strongest cooperation thanks to the contributions from the groups of Prof. Brillas in the former and Oturan in the latter.

Driven by the curiosity about the most productive authors in the field of the EF process, the co-occurrence network of the top 32 authors is depicted in Fig. 8b and Table 2. Despite Prof. Pignatello’s review endowed the strongest citation burst, he failed to appear in the list (Table 2), which is due to his research focus on non-electrochemical AOPs, including Fenton-based and persulfate-based methods. As the founder of the EF process, Prof. Brillas is the world-leading researcher both in number of publications and citations, along with Prof. Oturan, which is confirmed in their corresponding bigger circles Fig. 8b. A strong cooperation is also reflected by the numerous connection lines between authors. For example, researchers from both groups, like Prof. Sirés, Prof. Nihal Oturan and Prof. Garcia-Segura have further contributed to the EF process and are listed as the top 32 most productive authors in this area in Table 2. In other countries, we can see Prof. Minghua Zhou from China presenting a relatively big circle. Furthermore, close relationships between Profs. Zhou, Oturan and Brillas can be noticed as well.

| No | Reference | Citations | Title |
|----|-----------|-----------|-------|
| 1  | Pignatello (2006) [20] | 2628 | Advanced oxidation processes for organic contaminant destruction based on the Fenton reaction and related chemistry |
| 2  | Brillas (2009) [1] | 2176 | Electro-Fenton process and related electrochemical technologies based on Fenton's reaction chemistry |
| 3  | Martínez-Huitrle (2009) [110] | 1816 | Decontamination of wastewaters containing synthetic organic dyes by electrochemical methods: a general review |
| 4  | Moreira (2017) [112] | 1109 | Electrochemical advanced oxidation processes: a review on their application to synthetic and real wastewaters |
| 5  | Sirés (2014) [109] | 1172 | Electrochemical advanced oxidation processes: today and tomorrow. A review |
| 6  | Brillas (2015) [113] | 1245 | Decontamination of wastewaters containing synthetic organic dyes by electrochemical methods. An updated review |
| 7  | Bokare (2014) [114] | 1236 | Review of iron-free Fenton-like systems for activating H₂O₂ in advanced oxidation processes |
| 8  | Oturan (2014) [115] | 1146 | Advanced oxidation processes in water/wastewater treatment: principles and applications. A review |
| 9  | Martínez-Huitrle (2015) [111] | 941 | Single and coupled electrochemical processes and reactors for the abatement of organic water pollutants: a critical review |
| 10 | Sirés (2012) [108] | 698 | Remediation of water pollution caused by pharmaceutical residues based on electrochemical separation and degradation technologies: a review |
| 11 | Nidheesh (2012) [10] | 633 | Trends in electro-Fenton process for water and wastewater treatment: an overview |
| 12 | Nidheesh (2018) [116] | 520 | An overview on the removal of synthetic dyes from water by electrochemical advanced oxidation processes |

*Last access to the Web of Science database was on 1st August 2022*
Fig. 8  a Co-occurrence network of authors from the top 12 most cited publications. b Co-occurrence network of the top 32 most prolific authors in EF.
3 Conclusions

This bibliometrics-based review summarizes the development of the EF process in the past 20 years. Three main conclusions can be drawn:

(I) Eighty-three countries have contributed to the dramatic growth of publications on the EF process, with Profs. Brillas and Oturan as the leaders in this area.

(II) Four stages can be identified in the development of EF, and a general cognitive model including three perspectives has been proposed.

(III) The top 12 most cited articles, which correspond to review papers, and the top 32 productive authors in the field of EF process have been screened.

4 Future perspectives

According to the bibliometric trends in the EF process, the following future views are proposed:

(I) Gas-diffusion electrodes (GDEs) ensure a high oxygen concentration in the vicinity of the cathode and result in much higher current densities by orders of magnitude as compared to the submerged cathodes.
in EF [1]. However, the existence of a three-phase interface and its role in enhancing high current density in ORR, as most researchers claim, need direct evidence and further clarification. For example, a recent investigation on CO₂ electroreduction using GDEs suggested that the atomic liquid–solid two-phase is mainly responsible for the reactivity instead of the triple-phase [117]. In addition, challenges referred to water flooding and cathode corrosion during scale-up require further investigation both from a mechanistic and engineering point of view. Based on previous investigations [102, 118], the surface of carbon turns from hydrophobic to hydrophilic by oxygen functional group introduction even under the application of a negative potential (−0.4 V to −0.8 V vs RHE), suggesting some potential reasons for flooding. The role of each side, i.e., the gas-diffusion layer and the catalyst layer, needs to be clarified. For example, Nafion is widely used as the binder in the catalyst layer in GDEs used for EF process. Its role in enhancing reactant availability with hydrophobic (−CF₂) and hydrophilic (−SO₃⁻) functionalities has been mostly ignored [119].

(II) DFT is a useful tool to reveal the electronic effect in EF. However, there are still some discrepancies between the real interface and the constructed models for simulation. The supercell is built for calculation purposes and sometimes, only limited atoms are taken into consideration. Reactions in the cathode vicinity are dynamic and controlled by mass transport and activity/selectivity [37]. Moreover, the presence of cations in the double layer could tune the reactivity as well, which failed to be modeled in DFT simulation. Hence, a large-scale atomic/molecular massively parallel simulator (LAMMPS) should be used to simulate the complex reaction together with DFT in EF, thus offering a dynamic view. For example, a concerted theory-computation model proposed by Huang, which incorporated electronic effect, double-layer effect and mass transport, could be one alternative [120]. More importantly, the model design for DFT simulation is suggested to be validated prior to the calculation.

(III) In addition, operando spectroscopic or microscopic techniques combined with advanced electrochemical techniques are highly needed to analyze the dynamic electrode/electrolyte interface and cathode/catalyst structures under real electrochemical conditions, revealing the real-time evolution of cathode/catalyst since they may undergo dramatic changes under EF conditions [102, 119]. For example, in-situ X-ray photoelectron spectroscopy, in-situ electrochemical attenuated total reflection Fourier-transform infrared (ATR-FTIR) spectroscopy (ATR-FTIRs) or operando XAS measurement.

(IV) Electrode wettability plays a crucial role in H₂O₂ accumulation. For example, superaerophilic electrodes are transformed from underwater Wenzel–Cassie state with a gas pocket into water thoroughly wetted underwater Wenzel (UW) state [105] followed by H₂O₂ reduction. The evolution mechanism is still unclear and further clarification is required as well.

(V) The presence of cations like Na⁺, K⁺, Ca²⁺ and Mg²⁺ in wastewater may result in the cathodic salt precipitation caused by the electric field and local alkalization, as previously observed [37, 120]. This phenomenon could block the active sites and reduce the H₂O₂ generation even after acid-washing recovery [121]. It would definitely reduce its cathode lifespan. Hence, effective strategies should be put forward to alleviate salt precipitation, especially for scale-up applications. Aside from salt precipitation, it has been reported that some cations in the double layer might induce the cation promotion effect, consisting in the regulation of the rate-determining first electron transfer to O₂ following the order: Cs⁺ > K⁺ > Li⁺ [122–124]. However, the cation effect in the double layer within the context of EF process has not been investigated in detail yet.

(VI) Pulse electrolysis is a promising tool to pave the way to lower energy consumption. But its usage in the EF process is still at an early stage. Despite the fact that pulse method favors the in-situ regulation of active sites during 2e⁻-ORR [40] and mass transport of cathodic iron reduction [37], mechanistic research on a molecular level using a time-dependent continuum model is highly desirable, especially involved in the reconstruction of the electrode surface, reaction intermediates, the double-layer effect and mass transport effect.

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

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