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 micro, meso and macroscopic views, and future perspectives are discussed.

GRAPHICAL ABSTRACT

Hot topics for future research revealed by the bibliometric analysis

Co-occurrence network of the top keywords in the electro-Fenton process from 2002 to 2022 analyzed by VOSviewer

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**), H_2O_2 is formed via cathodic two-electron oxygen reduction reaction (2e$^-$-ORR) (**equation (1)**); for this, gaseous oxygen ($O_{2,gas}$) must be dissolved into the supporting electrolyte ($O_{2,bulk}$), 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 H_2O_2 diffuses away from the porous cathodic structure, undergoing catalytic decomposition upon Fenton's reaction with soluble iron ions (**equation (2)**), yielding the powerful oxidant hydroxyl radical ($E^0(\bullet OH|H_2O) = 2.8$ V vs. NHE) through the Haber-Weiss mechanism [2]. Unavoidably, H_2O_2 is partially reduced to H_2O 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 CO_2 under the action of $\bullet OH$, 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 (**equation (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 H_2 evolution (**equation (4)**) and the four-electron oxygen reduction reaction (4e$^-$-ORR, **equation (5)**) are typically the competing cathodic reactions that cause a decrease in current efficiency.
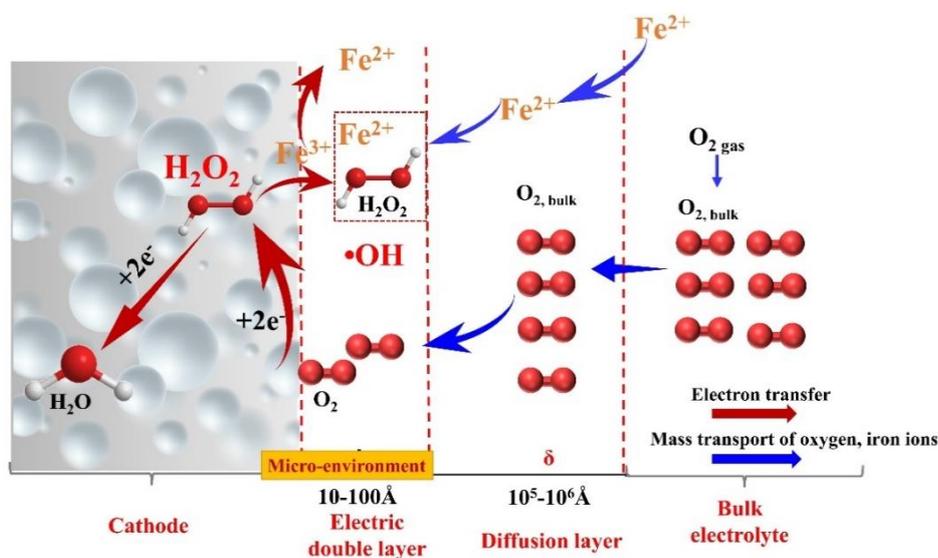

Fig. 1. A typical mechanism in the EF process involving in H_2O_2 formation and iron reduction.

The efficiency of EF is mainly dependent on the cathodic H_2O_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.

$$O_2 + 2e^- + 2H^+ \rightarrow H_2O_2 \tag{1}$$

$$H_2O_2 + Fe^{2+} \rightarrow Fe^{3+} + \bullet OH + OH^- \qquad \text{(Absolute rate constant } (k_1) \approx 70 \text{ M}^{-1}\text{ s}^{-1}) \tag{2}$$

$$Fe^{3+} + e^- \rightarrow Fe^{2+} \qquad (E^0 = 0.77 \text{ V vs. NHE}) \tag{3}$$

$$2H^+ + 2e^- \rightarrow H_2 \tag{4}$$

$$O_2 + 4e^- + 4H^+ \rightarrow 2H_2O \tag{5}$$

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 period 2002-2022 in the Web of Science Core Collection. 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, jus 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 Spain and Mexico. It can be noticed that Spanish research groups specialized in EF, with preponderance of the group from Profs. Brillas and Sirés, have a solid cooperation with France, Brazil, Mexico and Italy.
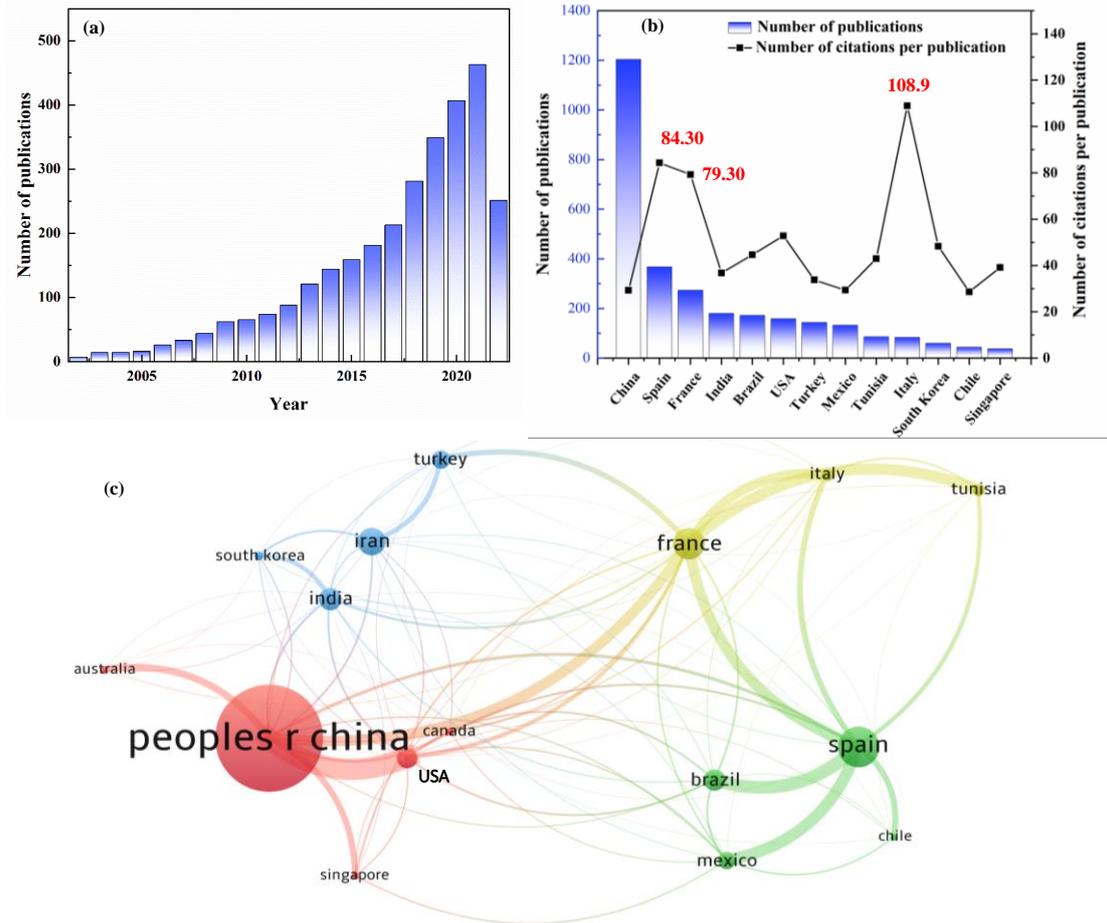
Fig. 2. (a) The number of publications between 2002 and 2022. (b) The first 13 most productive countries during the past 20 years along with the number of citations per publication. (c) Collaboration network between the top 15 most productive countries.

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.

Stage 1: Treatment of several pollutants
- Herbicides
- Synthetic dyes
- Leachate
- POPs
- Soil

Stage 2: Three emerging areas
- Novel cathode preparation
- Mineral iron catalyst
- Reactor design

Stage 4: Integral assessment
- Mechanism (ORR, iron reduction)-DFT
- Functional cathodes
- Heterogeneous catalysts
- Wider applications: e.g., disinfection

Stage 3: Mechanism investigations
- Oxygen reduction reaction for H_2O_2 generation
- Pulsed current to reduce H_2O_2 decomposition
- Heterogeneous catalysts

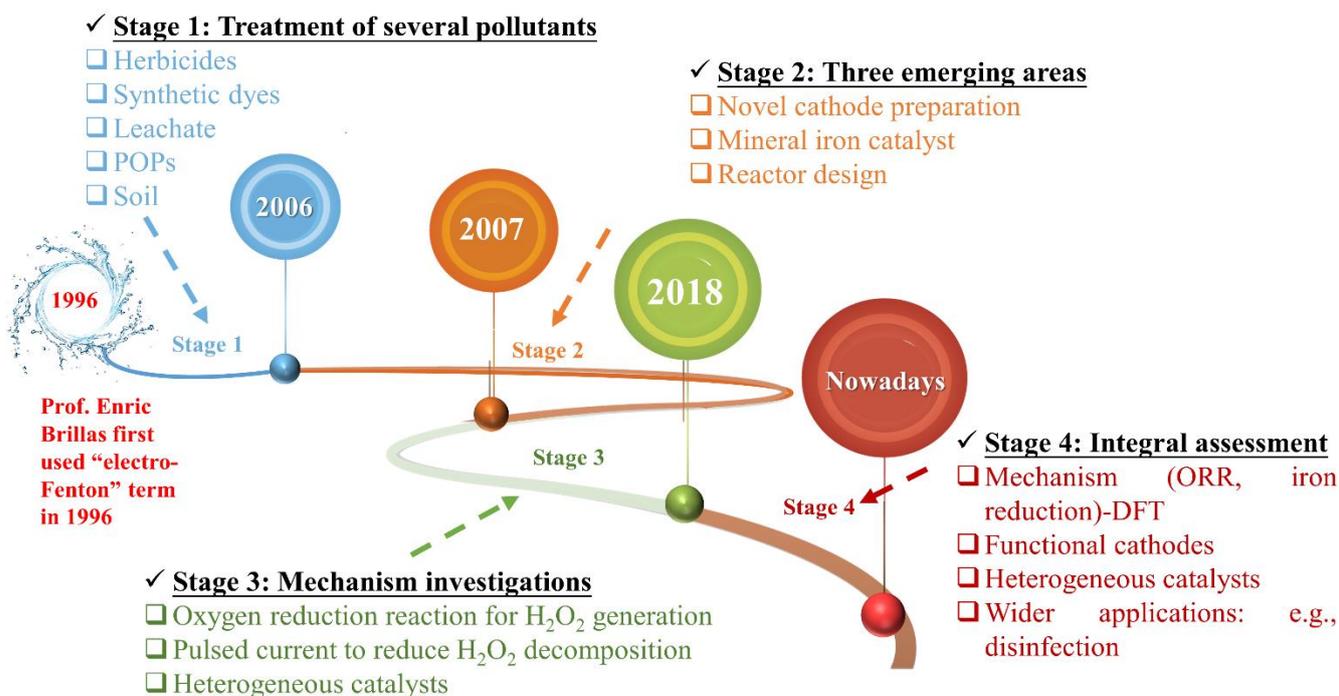Prof. Enric Brillas first used "electro-Fenton" term in 1996

Fig. 3. The four stages of the EF process development identified from the bibliometric analysis of 3012 research works published in the past two decades in the Web of Science Core Collection.

2.2.1 The first stage: treatment of several pollutants

The first stage dates back to 1996 when the term "electron-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

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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 Fe^{2+} 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 H_2O_2 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 H_2O_2 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 $Fe@Fe_2O_3$ nanowires-coated oxygen-diffusion electrodes [32].

(II) Mineral iron catalysts like goethite, alpha-FeOOH, magnetite (Fe_3O_4) 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

Fig. 4. Co-occurrence network of the top keywords with the most frequent appearance in articles on the EF process in 2018, the year opening the third stage of EF investigation.

The third stage of EF development was between 2007-2018, 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 H_2O_2 production in the context of EF process has not attracted great attention until 2018.

In fact, before 2018, the measured H_2O_2 concentration in EF publications was the accumulated one, according to **equation (6)** [37], which subtracts the decomposed H_2O_2 ($[H_2O_2]_{decomposed}$) from the generated one ($[H_2O_2]_{generated}$). Therefore, a decrease of the latter and a reduction of the former should boost the H_2O_2 accumulation. Regarding the methods to alleviate the H_2O_2 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 H_2O_2 [38].

The rate of $[H_2O_2]_{generated}$ via $2e^-$-ORR can be expressed as **equation (7)** [42], where k_{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 O_2 concentration will definitely boost the rate of $2e^-$-ORR for H_2O_2 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 H_2O_2 accumulation mainly by increasing the local oxygen concentration. But only local oxygen concentration enhancement is not enough for H_2O_2 boosting since both surface charge transfer and oxygen concentration determine the occurrence of the ORR, as indicated in **equation (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 (**equation (8)**), in which end-on adsorption of O_2 on active sites is desirable for the first electron transfer step to keep the O-O bond in $*OOH$, as deduced from Sabatier's principle for $2e^-$-ORR [46]. In addition, further dissociation steps of O-O bond in $*OOH$ to tune the desorption of $*OOH$ for H_2O_2 production rather than dissociation of O-O bonding with H_2O production should be guaranteed as well (**equations (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 CoS_2 nanoparticles [53]. Moreover, a series of focused reviews on cathodic H_2O_2 generation via $2e^-$-ORR has been published [6,7,54].

$$[H_2O_2]_{accumulated} = [H_2O_2]_{generated} - [H_2O_2]_{decomposed} \qquad (6)$$

$$k_{2e\text{-}ORR} = k_{het} \times [O_2] \qquad (7)$$

$$*O_2 + H^+ + e^- \rightarrow *OOH \qquad (8)$$

$$*OOH + H^+ + e^- \rightarrow H_2O_2 + * \qquad (9)$$

$$*OOH + H^+ + e^- \rightarrow *O + H_2O \qquad (10)$$

$$*OOH + * \rightarrow *O + *OH \qquad (11)$$

01
- ✓ Pyrite
- ✓ Magnetite
- ✓ Hematite
- ✓ Goethite
- ✓ Wüstite
- ✓ Lepidocrocite

Iron minerals

02
- ✓ MOF-based ZVI
- ✓ Zero-valent iron
- ✓ Iron foam

Zero-valent iron

03
- ✓ Organic polymers
  - Alginate, chitosan
- ✓ Inorganic materials
  - Activated carbon, alumina, graphene

Iron supported on synthetic structures

04
- ✓ Fly ash, industrial pyrite waste slag
- ✓ Bentonite
- ✓ Kaolinite
- ✓ Vermiculite

Iron supported on waste/soil

Fig. 5. The four types of heterogeneous catalysts commonly used in the EF process.

Apart from the ORR-to-H_2O_2 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 (FeS_2) **[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
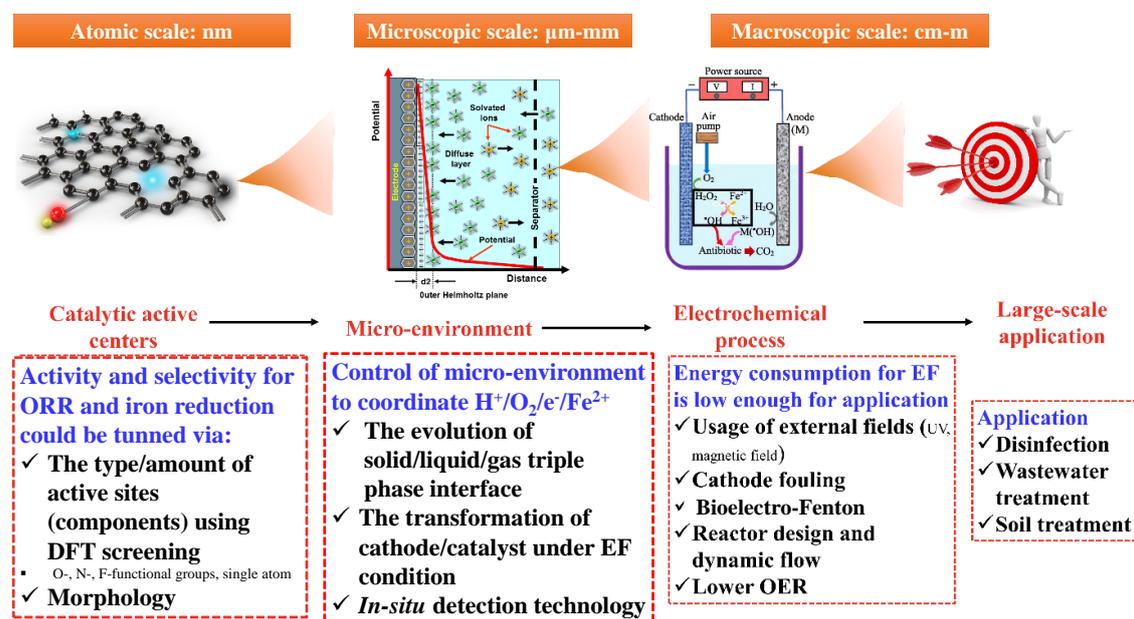
Fig. 6. A general cognitive model for the EF process.

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 (**equation (1)**) and iron reduction (**equation (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_2O_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_3O_4 catalyst **[91]**.

(II) The mesoscopic 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 transport 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, H_2O_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 modelled, 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].

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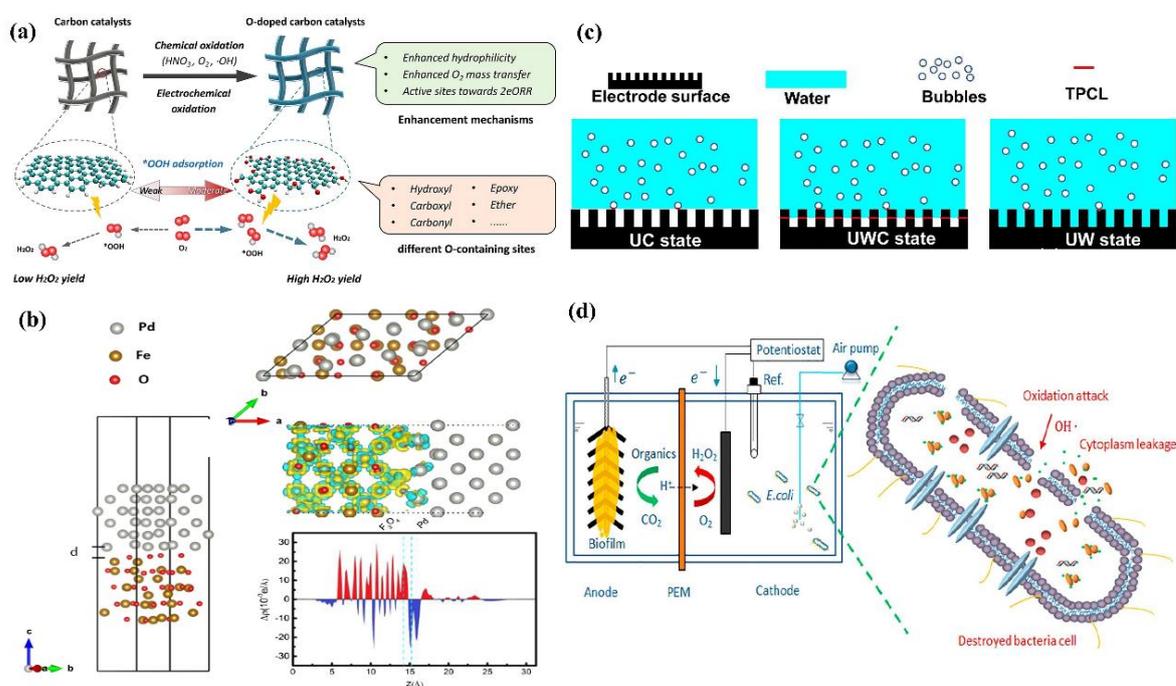
Fig. 7. (a) Doping with oxygen functional groups for ORR boosting [6]. (b) DFT calculations using Pd-Fe$_3$O$_4$ catalyst (adapted from [91]). (c) Various wetting states of UC, UWC and UW (adapted from [105]). (d) Disinfection mechanism (adapted from [106]).

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. On the other hand, Prof. Martínez-Huitle from Brazil has focused his work on electrocatalytic materials and electro-oxidation process. Two of his co-authored reviews **[110,111]** have arisen great attention since then. The strong cooperation between the authors of the most cited articles is also evidenced in **Fig. 8**.

Driven by the curiosity about the most productive authors in the field of the EF process, the co-occurrence network of the top 33 authors are 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 notices as well.
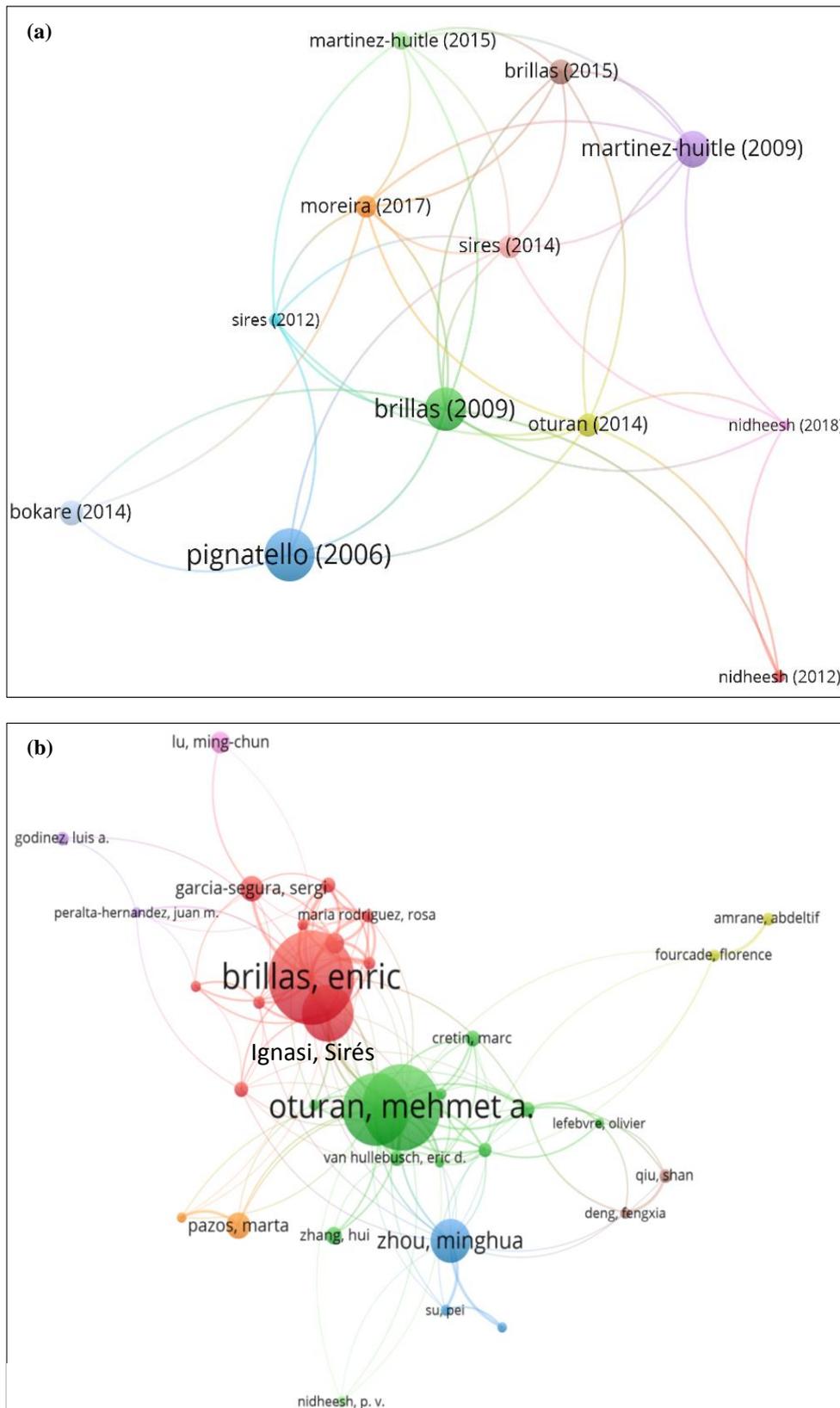
Fig. 8. (a) Co-occurrence network of authors from the top 12 most cited publications. **(b)** Co-occurrence network of the top 32 most prolific authors in EF.

Table. 1. The top 12 most cited publications on the EF process in the period 2012-2022

No	Reference	Citations[a]	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-Huitle (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 H_2O_2 in advanced oxidation processes
8	Oturan (2014) [115]	1146	Advanced oxidation processes in water/wastewater treatment: principles and applications. a review
9	Martínez-Huitle (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) [116]	633	Trends in electro-Fenton process for water and wastewater

				treatment: An overview
12	Nidheesh (2018) [117]	520		An overview on the removal of synthetic dyes from water by electrochemical advanced oxidation processes

Table. 2 The top 32 authors with a higher number of publications on the EF process

No	Author	Number of publications	Citations	Citation per publication	Institution	Country
1	Enric Brillas	174	19770	113.62	University of Barcelona	Spain
2	Mehmet A.Oturan	159	17321	99.55	Université Gustave Eiffel	France
3	Nihal Oturan	134	9597	55.16	Université Gustave Eiffel	France
4	Ignasi Sirés	105	10705	61.52	University of Barcelona	Spain
5	Minghua Zhou	82	4923	28.29	Nankai University	China
6	Marta Pazos	70	1881	10.81	University of Vigo	Spain
7	Sergi Garcia-Segura	48	3263	18.75	Arizona State University	USA
8	Ming Chun Lu	40	1303	7.49	National Chung Hsing University	China
9	Hui Zhang	33	1934	11.11	Wuhan University	China
10	Marc Cretin	30	1978	11.37	University of Montpellier	France

11	Carlos A. Martínez-Huitle	29	5170	29.71	Federal University of Rio Grande do Norte	Brazil
12	Hugo Olvera-Vargas	29	1139	6.55	Universidad Nacional Autónoma de México	Mexico
13	Eric D. van Hullebusch	28	2399	13.79	Paris City University	France
14	Emmanuel Mousset	27	1164	6.69	Université de Lorraine	France
15	Luis A. Godínez.	25	712	4.09	Universidad Autónoma de Querétaro	Mexico
16	Alireza Khataee	25	840	4.83	University of Tabriz	Iran
17	Shan Qiu	25	456	2.62	Harbin Institute of Technology	China
18	Abdeltif Amrane	24	694	3.99	Université de Rennes	France
19	Abdoulaye Thiam	24	945	5.43	Universidad Tecnológica Metropolitana	Chile
20	Florence Fourcade	23	688	3.95	Université de Rennes	France
21	Xie Quan	23	476	2.74	Dalian University of Technology	China
22	Fengxia Deng	22	456	2.62	Harbin Institute of Technology	China
23	Giovanni Esposito	22	2164	12.44	University of Milan	Italy

24	P.V. Nidheesh	22	2149	12.35	National Environmental Engineering Research Institute	India
25	Ricardo Salazar	22	824	4.74	University of Santiago	Chile
26	Salah Ammar	21	1594	9.16	Université de Gabès	Tunisia
27	María Ángeles Sanromán	21	744	4.28	University of Vigo	Spain
28	Pei Su	21	490	2.82	Nankai University	China
29	Olivier Lefebvre	20	491	2.82	National University of Singapore	Singapore
30	Juan M. Peralta-Hernández	20	823	4.73	Universidad de Guanajuato	Mexico
31	Clément Trellu	20	657	3.78	Université Paris-Est Marne-la-Vallée	France
32	Qizhan Zhang	20	350	2.01	Nankai University	China

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 CO_2 electroreduction using GDEs suggested that the atomistic liquid-solid two-phase is mainly responsible for the reactivity instead of the triple-phase [118]. 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 [102,119], 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 ($-CF_2$) and hydrophilic ($-SO_3^-$) functionalities has been mostly ignored [120].

(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. In fact, 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 [121]. 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 [102,120]. For example, *in-situ* X-ray photoelectron spectroscopy, *in-situ* electrochemical attenuated total reflection Fourier-transform infrared spectroscopy (ATR-FTIRS) or operando XAS measurement.

(IV) Electrode wettability plays a crucial role in H_2O_2 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 H_2O_2 reduction. The evolution mechanism is still unclear and further clarification is required as well.

(V) The presence of cations like Na^+, K^+, Ca^{2+} and Mg^{2+} in wastewater may result in the cathodic salt precipitation caused by the electric field and local alkalization, as previously observed [37,121]. This phenomenon could block the active sites and reduce the H_2O_2 generation even after acid-washing recovery [122]. 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 O_2 following the order: $Cs^+ >$ $K^+ > Li^+$ [123-125]. 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.

References:

[1] E. Brillas, I. Sirés, M.A. Oturan (2009) Electro-Fenton process and related electrochemical technologies based on Fenton's reaction chemistry. Chem Rev 109: 6570-6631. https://doi.org/10.1021/cr900136g

[2] S. Qiu, W. Tang, S. Yang, J. Xie, D. Yu, O. Garcia-Rodriguez, J. Qu, S. Bai, F. Deng (2022) A microbubble-assisted rotary tubular titanium cathode for boosting Fenton's reagents in the electro-Fenton process. J Hazard Mater 424: 127403. https://doi.org/10.1016/j.jhazmat.2021.127403

[3] J. Qu, X. Tian, X. Zhang, J. Yao, J. Xue, K. Li, B. Zhang, L. Wang, Y. Zhang (2022) Free radicals-triggered reductive and oxidative degradation of highly chlorinated compounds via regulation of heat-activated persulfate by low-molecular-weight organic acids. Appl Catal B: Environ 310: 121359. https://doi.org/10.1016/j.apcatb.2022.121359

[4] F. Deng, H. Olvera-Vargas, O. Garcia-Rodriguez, S. Qiu, F. Ma, Z. Chen, O. Lefebvre (2020) Unconventional electro-Fenton process operating at a wide pH range with Ni foam cathode and tripolyphosphate electrolyte. J Hazard Mater 396: 122641. https://doi.org/10.1016/j.jhazmat.2020.122641

[5] M. Mazzucato, C. Durante (2022) Insights on oxygen reduction reaction to H_2O_2: the role of functional groups and textural properties on the activity and selectivity of doped carbon electrocatalysts. Curr Opinion Electrochem 35: 101051. https://doi.org/10.1016/j.coelec.2022.101051

[6] W. Zhou, L. Xie, J. Gao, R. Nazari, H. Zhao, X. Meng, F. Sun, G. Zhao, J. Ma (2021) Selective H_2O_2 electrosynthesis by O-doped and transition-metal-O-doped carbon cathodes via O_2 electroreduction: A critical review. Chem Eng J 410: 128368. https://doi.org/10.1016/j.cej.2020.128368

[7] Y. Ding, W. Zhou, J. Gao, F. Sun, G. Zhao (2021) H_2O_2 electrogeneration from O_2 electroreduction by N-doped carbon materials: a mini-review on preparation methods, selectivity of N Sites, and prospects. Adv Mater Interfaces 8: 2002091. https://doi.org/10.1002/admi.202002091

[8] G. Divyapriya, P.V. Nidheesh (2020) Importance of graphene in the electro-Fenton process. ACS Omega 5: 4725-4732. https://doi.org/10.1021/acsomega.9b04201

[9] T. Yu, C.B. Breslin (2020) Graphene-modified composites and electrodes and their potential applications in the electro-Fenton process. Materials 13: 2254. https://doi.org/10.3390/ma13102254

[10] P.V. Nidheesh, R. Gandhimathi (2012) Trends in electro-Fenton process for water and wastewater treatment: An overview. Desalination 299: 1-15. https://doi.org/10.1016/j.desal.2012.05.011

[11] M.A. Oturan (2021) Outstanding performances of the BDD film anode in electro-Fenton process: applications and comparative performance. Curr Opinion Solid State Mater Sci 25: 100925. https://doi.org/10.1016/j.cossms.2021.100925

[12] V. Poza-Nogueiras, E. Rosales, M. Pazos, M.Á. Sanromán (2018) Current advances and trends in electro-Fenton process using heterogeneous catalysts - a review. Chemosphere 201: 399-416. https://doi.org/10.1016/j.chemosphere.2018.03.002

[13] A. Fernandes, M.J. Pacheco, L. Ciríaco, A. Lopes (2015) Review on the electrochemical processes for the treatment of sanitary landfill leachates: present and future. Appl Catal B: Environ 176-177: 183-200. https://doi.org/10.1016/j.apcatb.2015.03.052

[14] D. Syam Babu, P.V. Nidheesh (2021) A review on electrochemical treatment of arsenic from aqueous medium. Chem Eng Commun 208: 389-410. https://doi.org/10.1080/00986445.2020.1715956

[15] X. Zhang, Y. Zhang, Y. Wang, B.D. Fath (2021) Research progress and hotspot analysis for reactive nitrogen flows in macroscopic systems based on a CiteSpace analysis. Ecol Model 443: 109456. https://doi.org/10.1016/j.ecolmodel.2021.109456

[16] S.A. Ismail, W.L. Ang, A.W. Mohammad (2021) Electro-Fenton technology for wastewater treatment: a bibliometric analysis of current research trends, future perspectives and energy consumption analysis. J Water Process Eng 40: 101952. https://doi.org/10.1016/j.jwpe.2021.101952

[17] X. Ding, Z. Yang (2022) Knowledge mapping of platform research: a visual analysis using VOSviewer and CiteSpace. Electron Commer Res 22: 787-809. https://doi.org/10.1007/s10660-020-09410-7

[18] E. Brillas, E. Mur, J. Casado (1996) Iron(II) catalysis of the mineralization of aniline using a carbon- PTFE O_2- fed cathode. J Electrochem Soc 143: 49-53. http://dx.doi.org/10.1149/1.1836528

[19] G. Kaichouh, N. Oturan, M.A. Oturan, K. El Kacemi, A. El Hourch (2004) Degradation of the herbicide imazapyr by Fenton reactions. Environ Chem Lett 2: 31-33. https://doi.org/10.1007/s10311-004-0060-0

[20] J.J. Pignatello, E. Oliveros, A. MacKay (2006) Advanced oxidation processes for organic contaminant destruction based on the Fenton reaction and related chemistry. Crit Rev Env Sci Tec 36: 1-84. https://doi.org/10.1080/10643380500326564

[21] E. Guivarch, S. Trevin, C. Lahitte, M.A. Oturan (2003) Degradation of azo dyes in water by Electro-Fenton process. Environ Chem Lett 1: 38-44. https://doi.org/10.1007/s10311-002-0017-0

[22] J. Chen, M. Liu, J. Zhang, Y. Xian, L. Jin (2003) Electrochemical degradation of bromopyrogallol red in presence of cobalt ions. Chemosphere 53: 1131-1136. https://doi.org/10.1016/S0045-6535(03)00581-2

[23] H.Y. Lei, T. Bai, C.M. Wang, G.W. Yu (2004) Testing research on the treatment of dye wastewater by electro-Fenton reagent, in the 1st Symposium of the Chinese-German Forum on Geoenvironmental Engineering. 127-132. https://doi.org/10.1016/j.jelechem.2007.12.005

[24] H. Zhang, H.J. Choi, C.P. Huang (2005) Landfill leachate treatment by Fenton's reagent. The variation of leachate characteristics. Fresen Environ Bull 14: 1178-1183. https://doi.org/10.1016/10.1080/19443994.2013.778795

[25] Q. Xie, N. Lu, L. Wang, X. Lu (2005) Oxidation of hexachlorbenzene by the direct Fenton and EF-FEOX method. Fresen Environ Bull 14: 894-899. https://doi.org/ 10.1080/10643380500326564

[26] X. Zhao, J. Chen, M. Guo, C. Li, N. Hou, S. Bai (2022) Constructed wetlands treating synthetic wastewater in response to day-night alterations: Performance and mechanisms. Chem Eng J 446: 137460. https://doi.org/10.1016/j.cej.2022.137460

[27] A. Kyriacou, K.E. Lasaridi, M. Kotsou, C. Balis, G. Pilidis (2005) Combined bioremediation and advanced oxidation of green table olive processing wastewater. Process Biochem 40: 1401-1408. https://doi.org/10.1016/j.procbio.2004.06.001

[28] E. Brillas (2020) A review on the photoelectro-Fenton process as efficient electrochemical advanced oxidation for wastewater remediation. treatment with UV light, sunlight, and coupling with conventional and other photo-assisted advanced

technologies. Chemosphere 250: 126198. https://doi.org/10.1016/j.chemosphere.2020.126198

[29] G.R. Agladze, G.S. Tsurtsumia, B.I. Jung, J.S. Kim, G. Gorelishvili (2007) Comparative study of hydrogen peroxide electro-generation on gas-diffusion electrodes in undivided and membrane cells. J Appl Electrochem 37: 375-383. https://doi.org/10.1007/s10800-006-9269-x

[30] M. Zhou, Q. Yu, L. Lei (2008) The preparation and characterization of a graphite-PTFE cathode system for the decolorization of C.I. Acid Red 2. Dyes Pigm 77: 129-136. https://doi.org/10.1016/j.dyepig.2007.04.002

[31] M. Zhou, Q. Yu, L. Lei, G. Barton (2007) Electro-Fenton method for the removal of methyl red in an efficient electrochemical system. Sep Purif Technol 57: 380-387. https://doi.org/10.1016/j.seppur.2007.04.021

[32] Z. Ai, T. Mei, J. Liu, J. Li, F. Jia, L. Zhang, J. Qiu (2007) $Fe@Fe_2O_3$ core-shell nanowires as an iron reagent. 3. Their combination with CNTs as an effective oxygen-fed gas diffusion electrode in a neutral electro-Fenton system. J Phys Chem C 111: 14799-14803. https://doi.org/10.1021/jp073617c

[33] C.M. Sánchez-Sánchez, E. Expósito, J. Casado, V. Montiel (2007) Goethite as a more effective iron dosage source for mineralization of organic pollutants by electro-Fenton process. Electrochem Commun 9: 19-24. https://doi.org/10.1016/j.elecom.2006.08.023

[34] C. Badellino, C.A. Rodrigues, R. Bertazzoli (2007) Oxidation of herbicides by in situ synthesized hydrogen peroxide and Fenton's reagent in an electrochemical flow reactor: study of the degradation of 2,4-dichlorophenoxyacetic acid. J Appl Electrochem 37: 451-459. https://doi.org/10.1007/s10800-006-9275-z

[35] H. Liu, C. Wang, Xiangzhong, X. Xuan, C. Jiang, H. Cui (2007) A novel electro-Fenton process for water treatment: reaction-controlled pH adjustment and performance assessment. Environ Sci Technol 41: 2937-2942. https://doi.org/10.1021/es0622195

[36] M. Raji, S.A. Mirbagheri, (2020) A global trend of Fenton-based AOPs focused on wastewater treatment: a bibliometric and visualization analysis. Water Pract Technol 16: 19-34. https://doi.org/10.2166/wpt.2020.099

[37] F. Deng, S. Li, Y. Cao, M.A. Fang, J. Qu, Z. Chen, S. Qiu (2020) A dual-cathode pulsed current electro-Fenton system: Improvement for H_2O_2 accumulation and Fe^{3+} reduction. J Power Sources 466: 228342. https://doi.org/10.1016/j.jpowsour.2020.228342

[38] F. Deng, H. Olvera-Vargas, O. Garcia-Rodriguez, Y. Zhu, J. Jiang, S. Qiu, J. Yang (2019) Waste-wood-derived biochar cathode and its application in electro-Fenton for sulfathiazole treatment at alkaline pH with pyrophosphate electrolyte. J Hazard Mater 377: 249-258. https://doi.org/10.1016/j.jhazmat.2019.05.077

[39] F. Deng, S. Li, M. Zhou, Y. Zhu, S. Qiu, K. Li, F. Ma, J. Jiang (2019) A biochar modified nickel-foam cathode with iron-foam catalyst in electro-Fenton for sulfamerazine degradation. Appl Catal B: Environ 256: 117796. https://doi.org/10.1016/j.apcatb.2019.117796

[40] Y. Ding, L. Xie, W. Zhou, F. Sun, J. Gao, C. Yang, G. Zhao, Y. Qin, J. Ma (2022) Pulsed electrocatalysis enables the stabilization and activation of carbon-based catalysts towards H_2O_2 production. Appl Catal B: Environ 316: 121688. https://doi.org/10.1016/j.apcatb.2022.121688

[41] W. Zhou, J. Gao, Y. Ding, H. Zhao, X. Meng, Y. Wang, K. Kou, Y. Xu, S. Wu, Y. Qin (2018) Drastic enhancement of H_2O_2 electro-generation by pulsed current for

ibuprofen degradation: Strategy based on decoupling study on H_2O_2 decomposition pathways. Chem Eng J 338: 709-718. https://doi.org/10.1016/j.cej.2017.12.152

[42] E. Madrid, J.P. Lowe, K.J. Msayib, N.B. McKeown, Q. Song, G.A. Attard, T. Düren, F. Marken (2019) Triphasic nature of polymers of intrinsic microporosity induces storage and catalysis effects in hydrogen and oxygen reactivity at electrode surfaces. ChemElectroChem 6: 252-259. https://doi.org/10.1002/celc.201800177

[43] Z. Zhang, H. Meng, Y. Wang, L. Shi, X. Wang, S. Chai (2018) Fabrication of graphene@graphite-based gas diffusion electrode for improving H_2O_2 generation in Electro-Fenton process. Electrochim. Acta 260: 112-120. https://doi.org/10.1016/j.electacta.2017.11.048

[44] W. Zhou, X. Meng, L. Rajic, Y. Xue, S. Chen, Y. Ding, K. Kou, Y. Wang, J. Gao, Y. Qin, A.N. Alshawabkeh (2018) "Floating" cathode for efficient H_2O_2 electrogeneration applied to degradation of ibuprofen as a model pollutant. Electrochem Commun 96: 37-41. https://doi.org/10.1016/j.elecom.2018.09.007

[45] J.F. Pérez, J. Llanos, C. Sáez, C. López, P. Cañizares, M.A. Rodrigo (2018) The pressurized jet aerator: A new aeration system for high-performance H_2O_2 electrolyzers. Electrochem Commun 89: 19-22. https://doi.org/10.1016/j.elecom.2018.02.012

[46] C.H. Choi, H.C. Kwon, S. Yook, H. Shin, H. Kim, M. Choi (2014) Hydrogen peroxide synthesis via enhanced two-electron oxygen reduction pathway on carbon-coated Pt surface. J Phys Chem C 118: 30063-30070. https://doi.org/10.1021/jp5113894

[47] Z. Pan, K. Wang, Y. Wang, P. Tsiakaras, S. Song (2018) In-situ electrosynthesis of hydrogen peroxide and wastewater treatment application: a novel strategy for

graphite felt activation. Appl Catal B: Environ 237: 392-400. https://doi.org/10.1016/j.apcatb.2018.05.079

[48] Y. Ko, H. Kim, M.G. Seid, K. Cho, J. Choi, W. Lee, S.W. Hong (2018) Ionic-liquid-derived nitrogen-doped carbon electrocatalyst for peroxide generation and divalent iron generation: its application for removal of aqueous organic compounds. ACS Sustain Chem Eng 6: 14857-14865. https://doi.org/10.1021/acssuschemeng.8b03383.

[49] J. Liang, D. Tang, L. Huang, Y. Chen, W. Ren, J. Sun (2018) High oxygen reduction reaction performance nitrogen-doped biochar cathode: a strategy for comprehensive utilizing nitrogen and carbon in water hyacinth. Bioresource Technol 267: 524-531. https://doi.org/10.1016/j.biortech.2018.07.085

[50] J. Zhang, W. Zhou, L. Yang, Y. Chen, Y. Hu (2018) Co-N-doped MoO_2 modified carbon felt cathode for removal of EDTA-Ni in electro-Fenton process. Environ Sci Pollut Res 25: 22754-22765. https://doi.org/10.1007/s11356-018-2373-8

[51] A. Kumar, K. Sharma, A.R. Dixit (2021) A review on the mechanical properties of polymer composites reinforced by carbon nanotubes and graphene. Carbon Lett 31: 149-165. https://doi.org/10.1007/s42823-020-00161-x

[52] Y. Yan, W.I. Shin, H. Chen, S. Lee, S. Manickam, S. Hanson, H. Zhao, E. Lester, T. Wu, C.H. Pang (2021) A recent trend: application of graphene in catalysis. Carbon Lett 31: 177-199. https://doi.org/10.1007/s42823-020-00200-7

[53] C. Ridruejo, F. Alcaide, G. Álvarez, E. Brillas, I. Sirés (2018) On-site H_2O_2 electrogeneration at a CoS_2-based air-diffusion cathode for the electrochemical degradation of organic pollutants. J Electroanal Chem 808: 364-371. https://doi.org/10.1016/j.jelechem.2017.09.010

[54] J.H. Kim, Y. Kim, S.H. Joo (2020) Electrocatalyst design for promoting two-electron oxygen reduction reaction: Isolation of active site atoms. Curr Opinion Electrochem 21: 109-116. https://doi.org/10.1016/j.coelec.2020.01.007

[55] P.V. Nidheesh, H. Olvera-Vargas, N. Oturan, M.A. Oturan (2018) Heterogeneous electro-Fenton process: principles and applications, in: M. Zhou, M.A. Oturan, I. Sirés, 3rd edn. Springer, Singapore, pp 85-110

[56] N. Barhoumi, N. Oturan, S. Ammar, A. Gadri, M.A. Oturan, E. Brillas (2017) Enhanced degradation of the antibiotic tetracycline by heterogeneous electro-Fenton with pyrite catalysis. Environ Chem Lett 15: 689-693. https://doi.org/10.1007/s10311-017-0638-y

[57] Z. Es'Haghzade, E. Pajootan, H. Bahrami, M. Arami (2017) Facile synthesis of Fe_3O_4 nanoparticles via aqueous based electro chemical route for heterogeneous electro-Fenton removal of azo dyes. J Taiwan Inst Chem E 71: 91-105. https://doi.org/10.1016/j.jtice.2016.11.015

[58] F. Ghanbari, A. Hassani, S. Wacławek, Z. Wang, G. Matyszczak, K.A. Lin, M. Dolatabadi (2021) Insights into paracetamol degradation in aqueous solutions by ultrasound-assisted heterogeneous electro-Fenton process: Key operating parameters, mineralization and toxicity assessment. Sep Purif Technol 266: 118533. https://doi.org/10.1016/j.seppur.2021.118533

[59] J. Meijide, M. Pazos, M.Á. Sanromán (2019) Heterogeneous electro-Fenton catalyst for 1-butylpyridinium chloride degradation. Environ Sci Pollut Res 26: 3145-3156. https://doi.org/10.1007/s11356-017-0403-6

[60] E. Expósito, C.M. Sánchez-Sánchez, V. Montiel (2007) Mineral iron oxides as iron source in electro-Fenton and photoelectro-Fenton mineralization processes. J Electrochem Soc 154: 116. http://dx.doi.org/10.1149/1.2744134

[61] G. Zhang, S. Wang, F. Yang (2012) Efficient adsorption and combined heterogeneous/homogeneous Fenton oxidation of amaranth using supported nano-FeOOH as cathodic catalysts. J Phys Chem C 116: 3623-3634. https://doi.org/10.1021/jp210167b

[62] Y. Chen, L. Yang, J. Paul Chen, Y. Zheng (2019) Electrospun spongy zero-valent iron as excellent electro-Fenton catalyst for enhanced sulfathiazole removal by a combination of adsorption and electro-catalytic oxidation. J Hazard Mater 37: 576-585. https://doi.org/10.1016/j.jhazmat.2019.03.043

[63] S. Hu, J. Hu, B. Liu, D. Wang, L. Wu, K. Xiao, S. Liang, H. Hou, J. Yang (2018) In situ generation of zero valent iron for enhanced hydroxyl radical oxidation in an electrooxidation system for sewage sludge dewatering. Water Res 145: 162-171. https://doi.org/10.1016/j.watres.2018.08.027

[64] Z. Ye, J.A. Padilla, E. Xuriguera, E. Brillas, I. Sirés (2020) Magnetic MIL(Fe)-type MOF-derived N-doped nano-ZVI@C rods as heterogeneous catalyst for the electro-Fenton degradation of gemfibrozil in a complex aqueous matrix. Appl Catal B: Environ 266: 118604. https://doi.org/10.1016/j.apcatb.2020.118604

[65] Y. Zheng, S. Qiu, F. Deng, Y. Zhu, G. Li, F. Ma (2019) Three-dimensional electro-Fenton system with iron foam as particle electrode for folic acid wastewater pretreatment. Sep Purif Technol 224: 463-474. https://doi.org/10.1016/j.seppur.2019.05.054

[66] F. Deng, O. Garcia-Rodriguez, H. Olvera-Vargas, S. Qiu, O. Lefebvre, J. Yang (2018) Iron-foam as a heterogeneous catalyst in the presence of tripolyphosphate electrolyte for improving electro-Fenton oxidation capability. Electrochim Acta 272: 176-183. https://doi.org/10.1016/j.electacta.2018.03.160

[67] S. Barreca, J.J.V. Colmenares, A. Pace, S. Orecchio, C. Pulgarin (2014) Neutral solar photo-Fenton degradation of 4-nitrophenol on iron-enriched hybrid montmorillonite-alginate beads (Fe-MABs). J Photochem Photobiol A 282: 33-40. https://doi.org/10.1016/j.jphotochem.2014.02.008

[68] N. Taheri Ashtiani, B. Ayati (2022) Using chitosan-based heterogeneous catalyst for degradation of Acid Blue 25 in the effective electro-Fenton process with rotating cathodes. J Electroanal Chem 905: 115983. https://doi.org/10.1016/j.jelechem.2021.115983

[69] T. Luo, H. Feng, L. Tang, Y. Lu, W. Tang, S. Chen, J. Yu, Q. Xie, X. Ouyang, Z. Chen (2020) Efficient degradation of tetracycline by heterogeneous electro-Fenton process using Cu-doped Fe@Fe$_2$O$_3$: Mechanism and degradation pathway. Chem Eng J 382: 122970. https://doi.org/10.1016/j.cej.2019.122970

[70] F. Görmez, Ö. Görmez, B. Gözmen, D. Kalderis (2019) Degradation of chloramphenicol and metronidazole by electro-Fenton process using graphene oxide-Fe$_3$O$_4$ as heterogeneous catalyst. J Environ Chem Eng 7: 102990. https://doi.org/10.1016/j.jece.2019.102990

[71] L. Bounab, O. Iglesias, E. González-Romero, M. Pazos, M. Ángeles Sanromán, (2015) Effective heterogeneous electro-Fenton process of *m*-cresol with iron loaded actived carbon. RSC Adv 5: 31049-31056. http://dx.doi.org/10.1039/C5RA03050A

[72] P. Cao, X. Quan, K. Zhao, S. Chen, H. Yu, J. Niu (2020) Selective electrochemical H$_2$O$_2$ generation and activation on a bifunctional catalyst for heterogeneous electro-Fenton catalysis. J Hazard Mater 382: 121102. https://doi.org/10.1016/j.jhazmat.2019.121102

[73] D. Zhang, K. Yin, Y. Tang, Y. Wei, H. Tang, Y. Du, H. Liu, Y. Chen, C. Liu (2022) Hollow sea-urchin-shaped carbon-anchored single-atom iron as dual-

functional electro-Fenton catalysts for degrading refractory thiamphenicol with fast reaction kinetics in a wide pH range. Chem Eng J 427: 130996. https://doi.org/10.1016/j.cej.2021.130996

[74] H. Zhuang, S. Shan, J. Guo, Y. Han, C. Fang (2017) Waste rice straw and coal fly ash composite as a novel sustainable catalytic particle electrode for strengthening oxidation of azo dyes containing wastewater in electro-Fenton process. Environ Sci Pollut Res 24: 27136-27144. https://doi.org/10.1007/s11356-017-0319-1

[75] M. Sun, X. Ru, L. Zhai (2015) In-situ fabrication of supported iron oxides from synthetic acid mine drainage: high catalytic activities and good stabilities towards electro-Fenton reaction. Appl Catal B: Environ 165: 103-110. https://doi.org/10.1016/j.apcatb.2014.09.077

[76] W. Sun, N. Yu, J. Chen, Z. Gu, J. Wei, Y. Yao (2022) Heterogeneous Ti/PbO$_2$-electro-Fenton degradation of aromatic methane dyes using industrial pyrite waste slag as catalyst. Environ Sci Pollut Res 29: 50218-50236. https://doi.org/10.1007/s11356-022-19372-y

[77] M. Rostamizadeh, A. Jafarizad, S. Gharibian (2018) High efficient decolorization of Reactive Red 120 azo dye over reusable Fe-ZSM-5 nanocatalyst in electro-Fenton reaction. Sep Purif Technol 192: 340-347. https://doi.org/10.1016/j.seppur.2017.10.041

[78] O. Iglesias, M.A. Fernández De Dios, M. Pazos, M.A. Sanromán (2013) Using iron-loaded sepiolite obtained by adsorption as a catalyst in the electro-Fenton oxidation of Reactive Black 5. Environ Sci Pollut Res 20: 5983-5993. https://doi.org/10.1007/s11356-013-1610-4

[79] N. Qiao, H. Ma, M. Hu (2015) Design of a neutral three-dimensional electro-Fenton system with various bentonite-based Fe particle electrodes: a comparative

study, Mater Res Inno 19: 2-137. https://doi.org/10.1179/1432891715Z.0000000001315

[80] W. He, Q. Ma, J. Wang, J. Yu, W. Bao, H. Ma, A. Amrane (2014) Preparation of novel kaolin-based particle electrodes for treating methyl orange wastewater. Appl Clay Sci 99: 178-186. https://doi.org/10.1016/j.clay.2014.06.030

[81] J. Cheng, Z. Lu, X. Zhao, X. Chen, Y. Zhu, H. Chu (2021) Electrochemical performance of porous carbons derived from needle coke with different textures for supercapacitor electrode materials. Carbon Lett 31: 57-65. https://doi.org/10.1007/s42823-020-00149-7

[82] W.H. Danial, N.A. Norhisham, A.F. Ahmad Noorden, Z. Abdul Majid, K. Matsumura, A. Iqbal (2021) A short review on electrochemical exfoliation of graphene and graphene quantum dots. Carbon Lett 31: 371-388. https://doi.org/10.1007/s42823-020-00212-3

[83] S.Y. Lee, H.W. Jang, H.R. Lee, H. Joh (2021) Size effect of metal-organic frameworks with iron single-atom catalysts on oxygen-reduction reactions. Carbon Lett 31: 1349-1355. https://doi.org/10.1007/s42823-021-00292-9

[84] J. Bae, J. Hong (2021) Fabrication of nitrogen-doped porous carbon nanofibers for heavy metal ions removal. Carbon Lett 31: 1339-1347. https://doi.org/110.1007/s42823-021-00291-w

[85] S. Vargheese, D. Pattappan, K.V. Kavya, M.S. Sivaramkumar, R.T.R. Kumar, Y. Haldorai (2021) Heteroatom-doped mesoporous carbon prepared from a covalent organic framework/α-MnO_2 composite for high-performance supercapacitor. Carbon Lett 31: 1309-1316. https://doi.org/10.1007/s42823-021-00268-9

[86] M. Ramesh, L. Rajeshkumar, R. Bhoopathi (2021) Carbon substrates: a review on fabrication, properties and applications. Carbon Lett 31: 557-580. https://doi.org/10.1007/s42823-021-00264-z

[87] M. Zhao, Z. Cui, L. Fu, F. Ndayisenga, D. Zhou (2021) Shewanella drive Fe(III) reduction to promote electro-Fenton reactions and enhance Fe inner-cycle. ACS ES&T Water 1: 613-620. https://doi.org/10.1021/acsestwater.0c00126

[88] Z. Tao, X. Xu, L. Bi (2021) Density functional theory calculations for cathode materials of proton-conducting solid oxide fuel cells: a mini-review. Electrochem Commun 129: 107072. https://doi.org/10.1016/j.elecom.2021.107072

[89] A.M. Torres, J.D. Correa (2021) First-principles calculation of volatile organic compound adsorption on carbon nanotubes: Furan as case of study. Carbon Lett 31: 1061-1070. https://doi.org/10.1007/s42823-020-00221-2

[90] M.H. Abdel-Aziz, E.Z. El-Ashtoukhy, M. Bassyouni, A.F. Al-Hossainy, E.M. Fawzy, S.M.S. Abdel-Hamid, M.S. Zoromba (2021) DFT and experimental study on adsorption of dyes on activated carbon prepared from apple leaves. Carbon Lett 31: 863-878. https://doi.org/10.1007/s42823-020-00187-1

[91] Y. Zhou, X. Liu, Y. Zhao, S. Luo, L. Wang, Y. Yang, M.A. Oturan, Y. Mu (2018) Structure-based synergistic mechanism for the degradation of typical antibiotics in electro-Fenton process using Pd-Fe$_3$O$_4$ model catalyst: Theoretical and experimental study. J Catal 365: 184-194. https://doi.org/10.1016/j.jcat.2018.07.006

[92] Z. Lu, W. Xu, J. Ma, Y. Li, X. Sun, L. Jiang (2016) Superaerophilic carbon-nanotube-array electrode for high-performance oxygen reduction reaction. Adv Mater 28: 7155-7161. https://doi.org/10.1002/adma.201504652

[93] J.F. Pérez, J. Llanos, C. Sáez, C. López, P. Cañizares, M.A. Rodrigo (2016) Electrochemical jet-cell for the in-situ generation of hydrogen peroxide. Electrochem Commun 71: 65-68. https://doi.org/10.1016/j.elecom.2016.08.007

[94] J.D. García-Espinoza, I. Robles, A. Durán-Moreno, L.A. Godínez (2021) Study of simultaneous electro-Fenton and adsorption processes in a reactor containing porous carbon electrodes and particulate activated carbon. J Electroanal Chem 895: 115476. https://doi.org/10.1016/j.jelechem.2021.115476

[95] O.M. Cornejo, I. Sirés, J.L. Nava (2022) Cathodic generation of hydrogen peroxide sustained by electrolytic O_2 in a rotating cylinder electrode (RCE) reactor. Electrochim Acta 404: 139621. https://doi.org/10.1016/j.electacta.2021.139621

[96] G. Ren, M. Zhou, M. Liu, L. Ma, H. Yang (2016) A novel vertical-flow electro-Fenton reactor for organic wastewater treatment. Chem Eng J 298: 55-67. https://doi.org/10.1016/j.cej.2016.04.011

[97] J.F. Pérez, J. Llanos, C. Sáez, C. López, P. Cañizares, M.A. Rodrigo (2019) On the design of a jet-aerated microfluidic flow-through reactor for wastewater treatment by electro-Fenton. Sep Purif Technol 208: 123-129. https://doi.org/10.1016/j.seppur.2018.04.021

[98] J. Casado (2019) Towards industrial implementation of Electro-Fenton and derived technologies for wastewater treatment: a review., J Environ Chem Eng 7: 102823. https://doi.org/10.1016/j.jece.2018.102823

[99] X. Li, S. Chen, I. Angelidaki, Y. Zhang (2018) Bio-electro-Fenton processes for wastewater treatment: advances and prospects. Chem Eng J 354: 492-506. https://doi.org/10.1016/j.cej.2018.08.052

[100] S. Li, T. Hua, F. Li, Q. Zhou (2020) Bio-electro-Fenton systems for sustainable wastewater treatment: mechanisms, novel configurations, recent advances, LCA and

challenges. an updated review. J Chem Technol Biotechnol 95: 2083-2097. https://doi.org/10.1002/jctb.6332

[101] X. Ma, T. Rao, M. Zhao, Z. Jia, G. Ren, J. Liu, H. Guo, Z. Wu, H. Xie (2022) A novel induced zero-valent iron electrode for in-situ slow release of Fe^{2+} to effectively trigger electro-Fenton oxidation under neutral pH condition: advantages and mechanisms. Sep Purif Technol 283: 120160. https://doi.org/10.1016/j.seppur.2021.120160

[102] J. An, N. Li, Y. Wu, S. Wang, C. Liao, Q. Zhao, L. Zhou, T. Li, X. Wang, Y. Feng (2020) Revealing decay mechanisms of H_2O_2-based electrochemical advanced oxidation processes after long-term operation for phenol degradation. Environ Sci Technol 54: 10916-10925. https://doi.org/10.1021/acs.est.0c03233.

[103] I. Robles, E. Becerra, J.A. Barrios, C. Maya, B. Jiménez, F.J. Rodríguez-Valadez, F. Rivera, J.D. García-Espinoza, L.A. Godínez (2020) Inactivation of helminth eggs in an electro-Fenton reactor: Towards full electrochemical disinfection of human waste using activated carbon. Chemosphere 250: 126260. https://doi.org/10.1016/j.chemosphere.2020.126260

[104] G. Ren, M. Zhou, P. Su, W. Yang, X. Lu, Y. Zhang (2019) Simultaneous sulfadiazines degradation and disinfection from municipal secondary effluent by a flow-through electro-Fenton process with graphene-modified cathode. J Hazard Mater 368: 830-839. https://doi.org/10.1016/j.jhazmat.2019.01.109

[105] G. Xia, Y. Tian, X. Yin, W. Yuan, X. Wu, Z. Yang, G. Yu, Y. Wang, M. Wu (2021) Maximizing electrochemical hydrogen peroxide production from oxygen reduction with superaerophilic electrodes. Appl Catal B: Environ 299: 120655. https://doi.org/10.1016/j.apcatb.2021.120655

[106] S. Zhou, S. Huang, X. Li, I. Angelidaki, Y. Zhang (2018) Microbial electrolytic disinfection process for highly efficient Escherichia coli inactivation. Chem Eng J 342: 220-227. https://doi.org/10.1016/j.cej.2018.02.090

[107] C. Chen (2006) CiteSpace II: Detecting and visualizing emerging trends and transient patterns in scientific literature. J Assoc Inf Sci Technol 57: 359-377. https://doi.org/10.1002/asi.20317

[108] I. Sirés, E. Brillas (2012) Remediation of water pollution caused by pharmaceutical residues based on electrochemical separation and degradation technologies: a review. Environ Int 40: 212-229. https://doi.org/10.1016/j.envint.2011.07.012

[109] I. Sirés, E. Brillas, M.A. Oturan, M.A. Rodrigo, M. Panizza (2014) Electrochemical advanced oxidation processes: today and tomorrow. a review. Environ Sci Pollut Res 21: 8336-8367. https://doi.org/10.1007/s11356-014-2783-1

[110] C.A. Martínez-Huitle, E. Brillas, (2009) Decontamination of wastewaters containing synthetic organic dyes by electrochemical methods: a general review. Appl Catal B: Environ 87: 105-145. https://doi.org/10.1016/j.apcatb.2008.09.017

[111] C.A. Martínez-Huitle, M.A. Rodrigo, I. Sirés, O. Scialdone (2015) Single and coupled electrochemical processes and reactors for the abatement of organic water pollutants: a critical review. Chem Rev 115: 13362-13407. https://doi.org/10.1021/acs.chemrev.5b00361

[112] F.C. Moreira, R.A.R. Boaventura, E. Brillas, V.J.P. Vilar (2017) Electrochemical advanced oxidation processes: a review on their application to synthetic and real wastewaters. Appl Catal B: Environ 202: 217-261. https://doi.org/10.1016/j.apcatb.2016.08.037

[113] E. Brillas, C.A. Martínez-Huitle (2015) Decontamination of wastewaters containing synthetic organic dyes by electrochemical methods. an updated review. Appl Catal B: Environ 166-167: 603-643. https://doi.org/10.1016/j.apcatb.2014.11.016

[114] A.D. Bokare, W. Choi (2014) Review of iron-free Fenton-like systems for activating H_2O_2 in advanced oxidation processes. J Hazard Mater 275: 121-135. https://doi.org/10.1016/j.jhazmat.2014.04.054

[115] M.A. Oturan, J. Aaron (2014) Advanced oxidation processes in water/wastewater treatment: principles and applications. a review. Crit Rev Env Sci Tec 44: 2577-2641. https://doi.org/10.1080/10643389.2013.829765

[116] P.V. Nidheesh, R. Gandhimathi (2012) Trends in electro-Fenton process for water and wastewater treatment: an overview. Desalination 299: 1-15. https://doi.org/10.1016/j.desal.2012.05.011

[117] P.V. Nidheesh, M. Zhou, M.A. Oturan (2018) An overview on the removal of synthetic dyes from water by electrochemical advanced oxidation processes. Chemosphere 197: 210-227. https://doi.org/10.1016/j.chemosphere.2017.12.195

[118] N.T. Nesbitt, T. Burdyny, H. Simonson, D. Salvatore, D. Bohra, R. Kas, W.A. Smith (2020) Liquid-solid boundaries dominate activity of CO_2 reduction on gas-diffusion electrodes. Acs Catal 10: 14093-14106. https://doi.org/10.1021/acscatal.0c03319

[119] C. Dinh, T. Burdyny, M.G. Kibria, A. Seifitokaldani, C.M. Gabardo, F.P. García De Arquer, A. Kiani, J.P. Edwards, P. De Luna, O.S. Bushuyev, C. Zou, R (2018) Quintero-Bermudez, Y. Pang, D. Sinton, E.H. Sargent. CO_2 electroreduction to ethylene via hydroxide-mediated copper catalysis at an abrupt interface. Science 360: 783-787. https://doi.org/10.1126/science.aas9100

[120] G. de Arquer FP, D. CT, et al (2020) CO_2 electrolysis to multicarbon products at activities greater than 1 A cm^{-2}. Science, 367: 661-666. https://doi.org/10.1126/science.aay4217

[121] L. Zhang, J. Cai, Y. Chen, J. Huang (2021) Modelling electrocatalytic reactions with a concerted treatment of multistep electron transfer kinetics and local reaction conditions. J Phys Condens Matter 33: 504002. http://dx.doi.org/10.1088/1361-648x/ac26fb

[122] I. Salmerón, I. Oller, K.V. Plakas, S. Malato (2021) Carbon-based cathodes degradation during electro-Fenton treatment at pilot scale: changes in H_2O_2 electrogeneration. Chemosphere 275: 129962. https://doi.org/10.1016/j.chemosphere.2021.129962

[123] L.C. Pardo Pérez, A. Arndt, S. Stojkovikj, I.Y. Ahmet, et al (2022) Determining structure-activity relationships in oxide derived Cu-Sn catalysts during CO_2 electroreduction using X-ray spectroscopy. Adv Energy Mater 12: 2103328. https://doi.org/10.1002/aenm.202103328

[124] J. Lee, J.S. Lim, G. Yim, H. Jang, S.H. Joo, Y.J. Sa (2021) Unveiling the cationic promotion effect of H_2O_2 electrosynthesis activity of O-doped carbons. Acs Appl Mater Inter13: 59904-59914. https://doi.org/10.1021/acsami.1c17727

[125] X. Zhang, X. Zhao, P. Zhu, Z. Adler, Z. Wu, Y. Liu, H. Wang (2022) Electrochemical oxygen reduction to hydrogen peroxide at practical rates in strong acidic media. Nat Commun 13: 2880. https://doi.org/10.1038/s41467-022-30337-0

State-of-the-art review and bibliometric analysis

on electro-Fenton process

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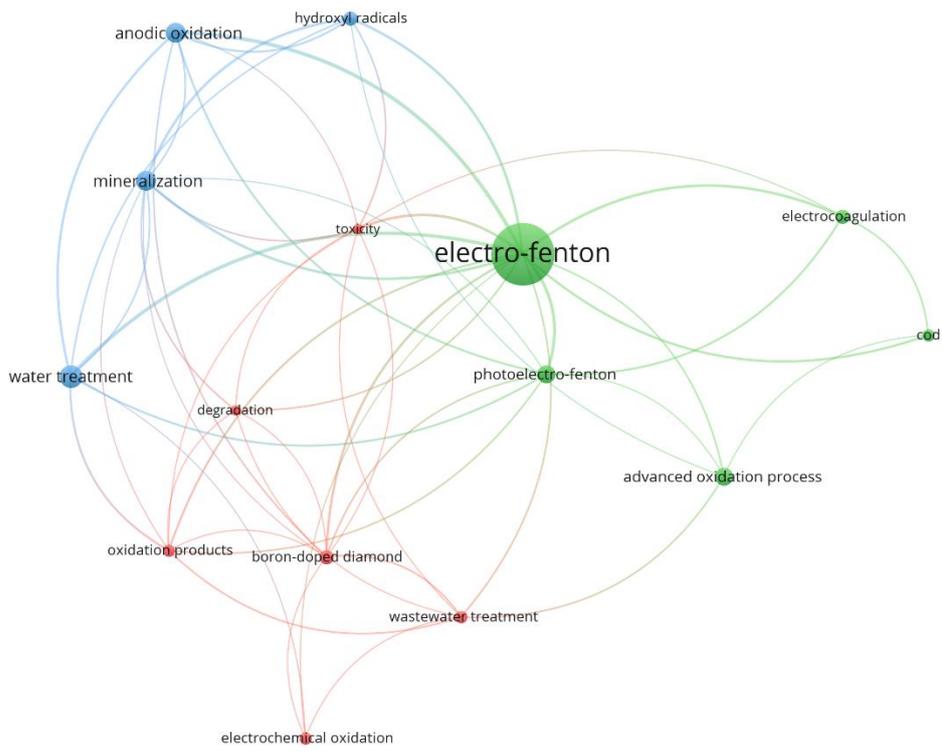
Fig. SI. 1. Co-occurrence network of the top keywords with the most frequent occurrence in articles on the EF process in 2014.
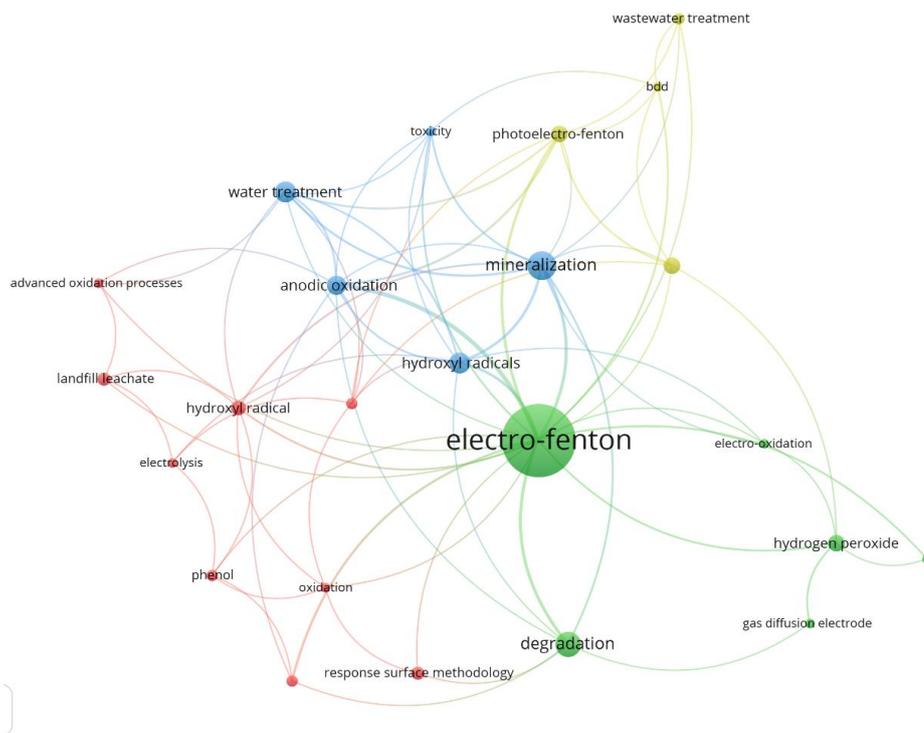
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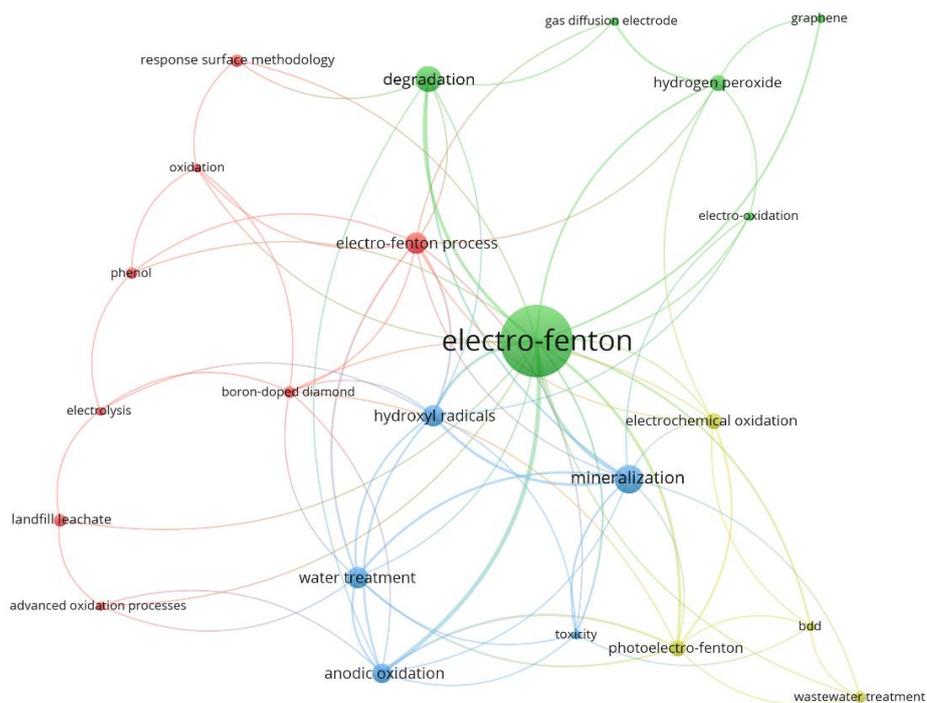

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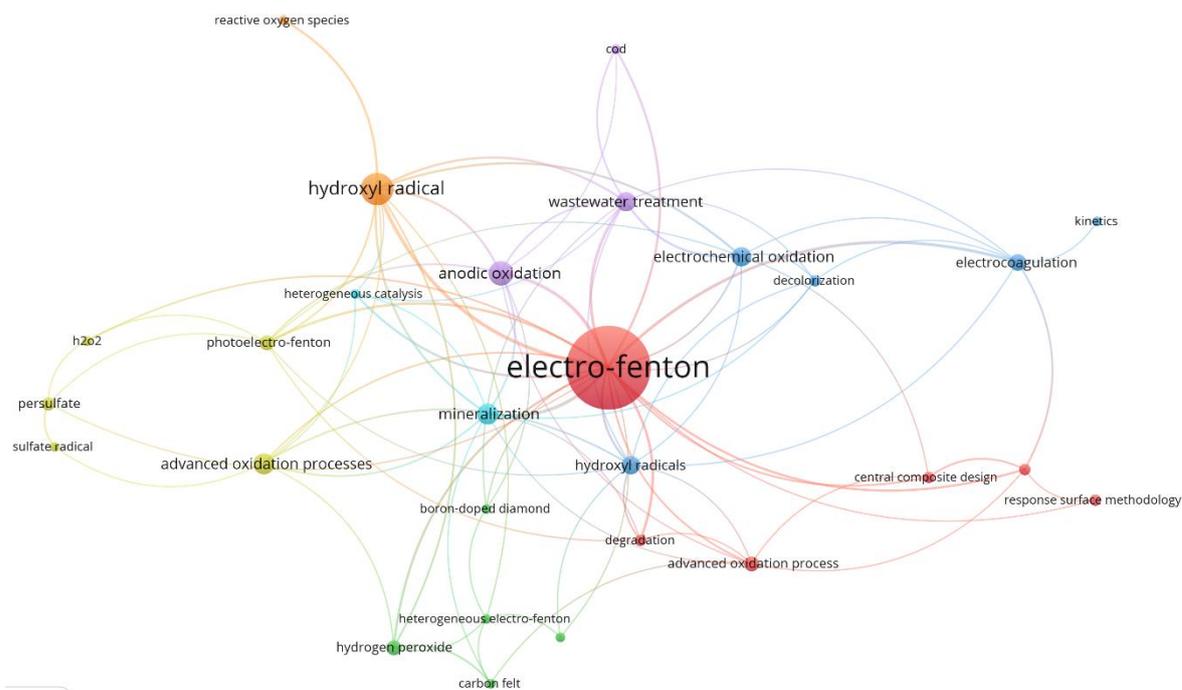
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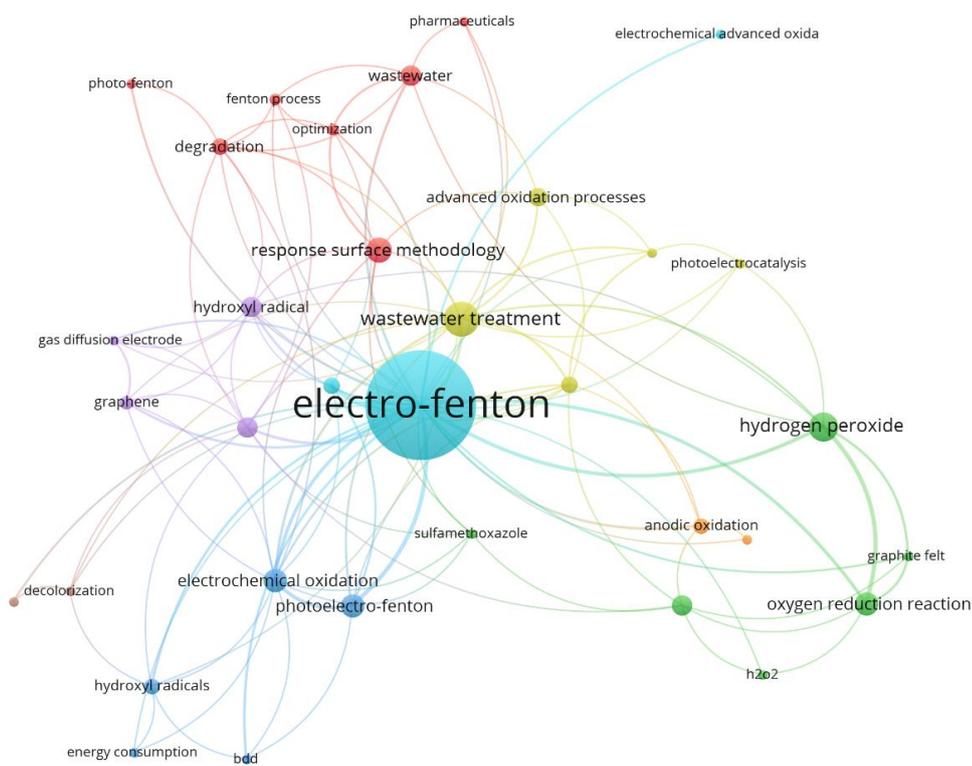

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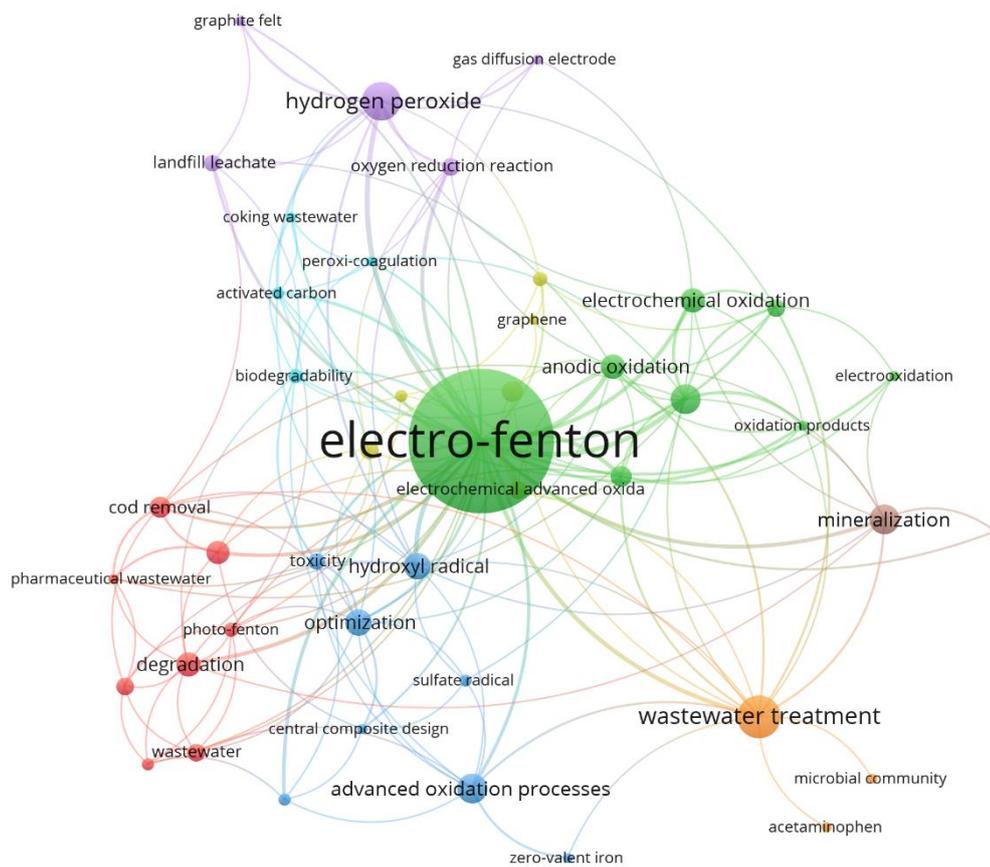
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Fig. SI. 7. Co-occurrence network of the top keywords with the most frequent occurrence in articles on the EF process in 2020.

Fig. SI. 8. Co-occurrence network of the top keywords with the most frequent occurrence in articles on the EF process in 2021.