



Fenton Technology for Wastewater Treatment: Dares and Trends

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Abstract

Fenton reaction remains an efficient technique for decomposing recalcitrant organic contaminants. Nevertheless, traditional Fenton response has many limitations like the necessity of acidic pH circumstance, the formation of iron sludge and the need for elevated chemical introductions. Procedures like heterogeneous Fenton, fluidized-bed Fenton, employment of chelating products and *in situ* formations of Fenton's reagent have been examined as likely solutions to such drawbacks. Bello *et al.* [1] presented an excellent discussion of the restrictions of Fenton reaction and the fresh manners for dealing with them and this work focuses on its main findings. The heterogeneous Fenton method stays the most largely examined thanks to the expansion achieved in catalysis. The fluidized-bed Fenton method has the capacity to diminish sludge formation and ameliorate technology efficiency. Chelating chemicals are employed to performing homogeneous Fenton at circumneutral pH, even if the potentially decisive impact of many chelating products remains a source of worry. *In situ* formation of Fenton's reagent via bio-electrochemical technique (bio-electro-Fenton) seems to be a likely manner to diminish the price related to Fenton's reagent. Despite the progress registered in the Fenton technologies, the classical process, and its ameliorated versions, membranes processes remain fundamental for secure wastewater treatment. As sure barriers towards pollution dispersal, processes such as nanofiltration should be coupled to Fenton techniques.

Subject Areas

Environmental Sciences

Keywords

Wastewater Treatment, Fenton Process, Pharmaceuticals and Personal Care

1. Introduction

There is an increasing endeavor to expand efficient techniques able of eliminating recalcitrant pollutants from wastewaters [1] [2]. Several organic contaminants, like pharmaceuticals and personal care products (PPCPs), are recalcitrant to the traditional wastewater treatment techniques [3] [4] [5]. Numerous investigations have proved the existence of diverse recalcitrant and emerging contaminants in the effluents of traditional wastewater treatment plants (WWTPs) [6] [7] [8]. Such contaminants might restrict the possible reuse of the treated wastewater and constitute dangers to public health and nature [9] [10] [11] [12] [13]. For instance, chronic toxicity and endocrine disruption have been related to the existence of even a low level of PPCP [14].

Because of the imperfections of traditional wastewater treatment techniques in dealing with recalcitrant contaminants [15] [16] [17] [18], attempts have been escalated to discover substitutional techniques. Even if physicochemical processes like adsorption might reduce recalcitrant contaminants from wastewater, they are mainly separation techniques and additional remediation may be needed [19] [20] [21]. Biodegrading recalcitrant contaminants are frequently slow and mainly transform the organic contaminants into several intermediates, which may therewith cumulate in nature [22] [23]. Consequently, efficient remediation necessitates the total mineralization of such recalcitrant contaminants [1] [24] [25]. This may be attained by advanced oxidation processes (AOPs) [26] [27] [28], which are capable of efficaciously oxidizing organic chemicals.

During AOPs, hydroxyl radicals ($\cdot\text{OH}$) are produced through a series of methods [29]. The $\cdot\text{OH}$ (oxidation potential = 2.8 eV) could enter in mutual actions with the contaminants (rate constant: $109 \text{ M}^{-1}\cdot\text{s}^{-1}$), oxidizing them to simpler intermediates and probably to CO_2 and H_2O [30]. During the last decade, AOPs have attracted huge attention. They are viewed as the preferable techniques for decomposing recalcitrant contaminants [31]. Usually, AOPs are categorized following the procedure of producing the $\cdot\text{OH}$. Such procedures comprise Fenton oxidation [30], photocatalysis [32], UV-founded techniques, ozonation [29], sonolysis [33] and electrochemical oxidation [1] [34].

Fenton oxidation is an efficacious AOP and implies a catalytic degradation of hydrogen peroxide (H_2O_2) by ferrous iron (Fe^{2+}) to form $\cdot\text{OH}$ following Equation (1) [18]. The produced $\cdot\text{OH}$ after that oxidizes the organic contaminant (Equation (2)). The initial step of the technique is a rapid degradation of H_2O_2 by Fe^{2+} [35], which forms a significant quantity of $\cdot\text{OH}$ and transform Fe^{2+} to Fe^{3+} [36] [37] (Equation (3)) [1]. Even if Fenton oxidation is mostly defined in such straightforward stages, the technology is considerably more combined since it implicates numerous additional reactions. Usually, such techniques may be

largely classified into initiation, propagation and termination reactions [38].



The $\cdot\text{OH}$ may enter in mutual reactions with organic matter and/or Fenton catalyst through either of three routes: 1) hydroxyl addition, 2) hydrogen abstraction or 3) electron transfer [39]. Hydroxyl addition happens with organic chemicals possessing aromatic systems or carbon-carbon multiple bonds (Equation (4)). Hydrogen abstraction takes place with unsaturated organic chemicals (Equation (5)) while electron transfer happens if $\cdot\text{OH}$ interacts with inorganic ions (Equation (6)) [1].



The most important benefits of Fenton process are the simplicity of the method and the efficient decomposition of contaminants. Further, both Fe^{2+} [40] and H_2O_2 are easily obtainable, simple to manipulate and environmentally harmless. Fenton oxidation has been largely tried for dealing with diverse wastewater streams like textiles [41] [42], pharmaceuticals [43] [44], olive mill [45] [46], leachate [47], agrochemicals [48] [49] and other recalcitrant pollutants. Moreover, Fenton oxidation was presented as an environmentally secure choice for sludge conditioning and treatment [50]. For instance, researchers [51] tried Fenton process for the conditioning of sewage sludge and examined the contribution of iron [52] species and pH optimization. The Fenton oxidation transformed most of the sludge-bound water to free water, improving its dewater-capacity. The process was more assisted by the coagulating impact of Fe^{3+} [1] [53] [54] [55] [56].

Fenton process is influenced by diverse running factors, comprising the level and ratio of Fenton's reagent (H_2O_2 and Fe^{2+}), solution pH, time, temperature, contaminant concentration and the type of the reaction matrix [1]. Nevertheless, Fenton's reagent and pH are frequently viewed as the most crucial factors. The level of H_2O_2 dictates the possible $\cdot\text{OH}$ that will be formed in the method. This implies that the more H_2O_2 , the more $\cdot\text{OH}$ will be possibly produced, following the level of Fe^{2+} and different working circumstances. Nevertheless, a surplus level of H_2O_2 may conduct to a scavenging impact where the H_2O_2 reacts with the $\cdot\text{OH}$, transforming it to the less reactive $\text{HO}_2\cdot$ [21]. Further, Fe^{2+} works as a catalyst for the degradation of H_2O_2 . Consequently, augmenting the level of Fe^{2+} may conduct a more important catalytic degradation of H_2O_2 . Nevertheless, overabundant quantities of Fe^{2+} may as well conduct to scavenging of $\cdot\text{OH}$, which will curb the technique efficiency [57]. Moreover, an elevated level of Fe^{2+} may as well conduct to an elevated sludge formation at the end of the method. As a re-

sult, the ratio of H_2O_2 to Fe^{2+} is a critical precaution to guarantee the optimum formation of $\cdot\text{OH}$ and avert the scavenging impact.

The wastewater pH is an additional crucial factor touching Fenton process. Generating $\cdot\text{OH}$ is governed by the pH of the reaction medium; indeed, more elevated pH conducts to the precipitation of Fe (III) in the form of iron hydroxideflocs [1] [58]. Therefore, the formation of $\cdot\text{OH}$ and the dependent decomposition of contaminants via the Fenton process is efficient below acidic pH, *i.e.*, in the interval of 2.8 - 3.5 [59] [60] [61]. In other words, the pH of the wastewater should be regulated inside this domain to guarantee ferrous and ferric irons with a view to preserve their catalytic capacity [62]. As mentioned above, the temperature was counted to possess an unfavorable influence on Fenton response. This was attributed to the worry for probable degradation of H_2O_2 above 90°C and the energy necessity [63]. Therefore, researches on Fenton methods have widely been performed below ambient temperature. Nevertheless, modern investigations have proved that augmented temperatures may elevate the oxidation rate and improve Fenton's response. For instance, scientists [64] noted an amelioration of the transformation of linear alkylbenzenesulfonate through Fenton reaction if the temperature was elevated from 20°C to 94°C . At the lower temperature, 61% transformation was reached, which augmented to 99.9% at the higher temperature. Many fresh investigations have as well shown the improvement of Fenton response at more increase temperature [65] [66]. The favorable impact of higher temperatures may be addressed to a more performant utilization of H_2O_2 , which conducts to the more important formation of $\cdot\text{OH}$ [67]. The type of reaction matrix stays a crucial factor as the existence of organic/inorganic chemicals in wastewater may lead to a scavenging impact on the $\cdot\text{OH}$. Mineral chemicals like NaCl , Na_2CO_3 , and Na_2SO_3 are detected in different levels in genuine wastewaters and may react with $\cdot\text{OH}$, diminishing their accessibility [68].

The Fenton process undergoes many restrictions that prevent its large-scale usages. One of such barrier remains the immoderate formation of iron sludge that causes troubles in the recycling and can lead to resultant contamination. Additional restrictions involve the need for a limited domain of working pH and elevated chemical inputs [69]. As a consequence, handling the disadvantages of the Fenton process constitutes an endless study attempt in the area of the Fenton method for recalcitrant wastewater treatment. Many of the procedures to deal with such restrictions comprise the employment of heterogeneous Fenton process [70] [71], fluidized-bed Fenton technique [72], and electro-Fenton [73].

2. Fenton Technology for Recalcitrant Wastewater Treatment: Review Papers' State of the Art

The domain of Fenton technology for recalcitrant wastewater treatment was the subject of numerous review papers. Such literature discussions focused on diverse features of the Fenton technique comprising the basis and usages of conventional Fenton oxidation, heterogeneous Fenton process, electro-Fenton with its related processes, fluidized-bed Fenton process, Fenton process at circum-

neutral pH employing chelating agents and hybrid techniques, where Fenton reaction is integrated with additional techniques. Several review papers embraced the large domain of AOPs, like those mentioned by Oturan and Aaron [74], in which the Fenton process is evaluated with different AOPs. Nevertheless, Bello *et al.* [1] in their work largely encompassed those reviews where Fenton oxidation is the main interest (see in [1] (their **Table 1**) for a summary of many of these reviews).

Neyens and Baeyens [30] presented an evaluative survey of the conventional Fenton process, concentrating on the kinetics, the impact of running factors, the importance of the ratio of Fenton's reagent and the usage of Fenton method in sludge dewatering. In the following publication, Pignatello *et al.* [75] offered a global discussion of the pathways of Fenton and Fenton-similar processes, reaction routes and intermediates, changed configurations of Fenton process (heterogeneous Fenton, photo-helped Fenton [76], etc.) and their usages to water and soil remediation. More lately, scientists [77] [78] displayed analysis of Fenton-similar technologies for organic wastewater remediation. They concentrated on running factors, changed Fenton-similar methods, catalysts, and economic considerations [1].

Brillas *et al.* [79] published a thorough discussion of the electro-Fenton and similar electrochemical techniques that are founded on Fenton's response. They focused on the basics of Fenton's chemistry, the progression of the electro-Fenton method, cell arrangement, working factors, pathways, integrated electro-Fenton techniques, and their ecological utilizations. Newly, Poza-Nogueiras *et al.* [80] suggested an analysis of the present tendencies and developments in electro-Fenton technology focusing on the implementation of heterogeneous catalysts. They concentrated on the diverse sorts of catalysts and elaboration procedures, the influence of running indicators, pathways and usages in treating water, soil and adsorbent renewal. Researchers [81] evaluated the basic concepts and usages of electro-Fenton technology. The review concentrated on the kinds of catalyst and functionalized cathodic materials, route of catalytic activation of H_2O_2 and setup arrangement. A discussion of the bio-electro-Fenton was lately published by Li *et al.* [82] [83] who focused on its basics, working factors, reactor conception, and economic features [1].

Table 1. Probable impacts of pH on Fenton process [1].

pH below optimum	Reference	pH above optimum	Reference
Scavenging of $\cdot OH$ by excess H^+	[88]	Deficiency of H^+ , which hampers $\cdot OH$ formation	[59]
Formation of $[Fe(H_2O)]^{2+}$ which has low oxidative power	[89]	Decomposition of H_2O_2	[90]
Prevent the interaction between Fe^{3+} and H_2O_2	[91]	Decline in the oxidative power of $\cdot OH$	[92]
H_2O_2 may be unstable to formed $H_3O_2^+$ which impedes production of $\cdot OH$	[93]	Precipitation of Fe^{3+} as iron hydroxide	[60] [61]

Even if such analyses have embraced the basics of Fenton reaction and identical methods, a discussion concentrating on the main restrictions of Fenton technology and the new development to treating them has not been given until the excellent work done by Bello *et al.* [1]. Indeed, they discussed the fresh advance to dealing with the main restrictions of Fenton's response for recalcitrant wastewater treatment. Primarily, they analyzed the restrictions of Fenton technology, pursued by a pertinent discussion of the modern references on probable procedures for dealing with such restrictions. For each action plan, they assessed the basics and exemplary implementations in decomposing recalcitrant organic contaminants. They also suggested viewpoints on potential orientations for the next research. In fact, Bello *et al.* [1]'s analysis remains appropriate because of the augmenting investigation works to resolving the restrictions of the Fenton technology.

3. Restrictions of Fenton Process and Procedure to Deal with Them

3.1. Fluidized-Bed Fenton Process

The Fenton process has many obstacles that prevent its industrial implementation. Fenton technique is greatly touched by the solution pH, which requires to be fixed in the acidic domain to avert the precipitation of Fe^{3+} into iron hydroxide ($\text{Fe}(\text{OH})_3(\text{s})$). **Table 1** recaps the impacts of pH outside the optimum domain. The necessity of delicate dominance of pH and the hardness of running in acidic circumstances stop the workable implementations of Fenton technology [84]. Bigger pH conducts to complexation reactions and precipitation of iron oxides, causing the formation of excessive sludge. As a result, sludge production constitutes one more inconvenient of the traditional Fenton process [85] [86]. Sludge formation opens the hazard of additional contamination and the necessity for sludge remediation and recycling. The necessity of supplementary remediation of sludge is an origin of worry since the cost of sludge remediation may be above to 35% - 50% of the total operating cost of the wastewater treatment [30]. More difficulties of the Fenton process involve elevated chemical utilization, instability of the Fenton's reagent, undesirable reactions and loss of oxidant, hardness in regulating the reagent concentrations and the exigency to neutralize the treated wastewater prior recycling [75] [87]. **Table 2** underlines the main restrictions of the Fenton technique and several of the likely procedures to deal with them [1].

There is increasing attention in investigations to resolve the restrictions of the traditional Fenton process. Not many steps have appeared as likely solutions as listed in **Table 2**. One such procedure remains the expansion of heterogeneous Fenton oxidation, in which iron oxides or other metal oxides are employed as heterogeneous catalysts [38]. Fluidized-bed Fenton technique is second planning, in which a fluidized-bed reactor is integrated with the homogeneous Fenton reaction to enhance process efficiency and decrease sludge formation. An additional manner implicates performing Fenton oxidation at near-neutral pH

Table 2. Main restrictions of classical Fenton process and their likely solutions [1].

S/N	Restriction	Causes	Probable solutions	References
1	Requirement of acidic pH (narrow range of operational pH)	<ul style="list-style-type: none"> • Production of $\cdot\text{OH}$ requires acidic pH (2.5 - 3.5) • Acidic pH is required to keep Fe^{2+} in soluble form • pH above 4 promotes complex formation and sludge generation 	<ul style="list-style-type: none"> • Use of heterogeneous catalyst (iron oxide, transition metal, composites, etc.) • Use of chelating agents such as EDTA, oxalate, etc. 	[62] [84] [94]
2	Excessive sludge generation	<ul style="list-style-type: none"> • Neutralization of the Fenton reaction 	<ul style="list-style-type: none"> • Fluidized-bed Fenton process • Heterogeneous Fenton • Electro-Fenton 	[69] [72] [95] [96]
3	Chemical consumption/cost of chemicals (H_2O_2)	<ul style="list-style-type: none"> • Large amount of H_2O_2 may be necessary to completely mineralize pollutants • Scavenging of H_2O_2 anions/cations present 	<ul style="list-style-type: none"> • Use of electro-Fenton/bio-electro-Fenton and other technologies with <i>in situ</i> generation of H_2O_2 • Other alternative oxidants 	[77] [78] [97]

via the introduction of a chelating chemical to produce iron-complexes [60] [61]. Manners able of *in situ* generation of oxidants (H_2O_2) like electro-Fenton and bio-electro-Fenton (microbial fuel cell) stay interesting techniques for treating the elevated chemical necessity. Bello *et al.* [1] analyzed such procedures below four classes: 1) fluidized-bed Fenton process, 2) heterogeneous Fenton processes, 3) Homogeneous Fenton at neutral pH using chelating agents and, 4) *in situ* production of Fenton's reagent.

Immoderate sludge formation is between the main imperfections of the conventional Fenton reaction [1]. Several scientists have tried the application of a fluidized-bed reactor to realize Fenton's response, the so-named fluidized-bed Fenton process. In the fluidized-bed Fenton process, solids carriers, such as SiO_2 (Figure 1), are employed to trigger the crystallization of iron oxide and diminish sludge production [95] [98]. The fluidization improves mixing and raises $\cdot\text{OH}$ -contaminant interaction. Further, recirculation gives more oxygen into the setup, which may improve the technique efficiency by keeping $\cdot\text{OH}$ [99].

The first investigation on the fluidized-bed Fenton process was published by Chou and Huang [100]. They examined the oxidation of benzoic acid. Huang and Huang [101] [102] applied Fenton oxidation for decomposing phenol in a fluidized-bed setup. In the same way, Anotai *et al.* [103] noted the decomposition of nitrobenzene and iron crystallization employing a fluidized-bed Fenton process [1]. Following researches were mainly oriented to sludge decrease and process efficiency in decomposing diverse recalcitrant contaminants like dimethyl sulfoxide [98], dyes [104], bisphenol A [105] and phthalocyanine (Figure 2) [106].

3.2. Electro-Fenton Process

Using electrochemical technology in the Fenton oxidation is rising as an encouraging option. In electrochemical technology, the electron is employed as the

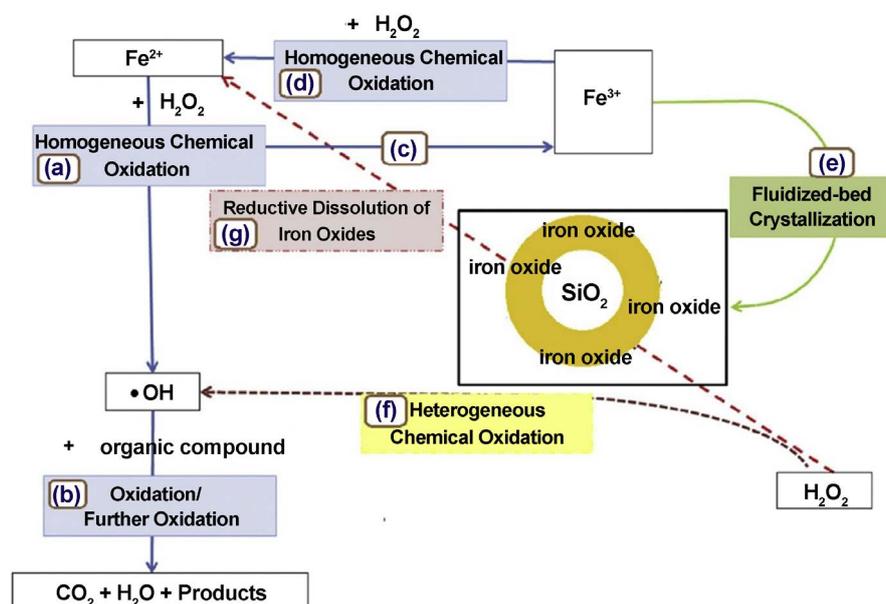


Figure 1. Various processes that occur in fluidized-bed Fenton process using SiO_2 as carriers, 1) homogeneous reaction between Fe^{2+} and H_2O_2 , 2) crystallization of Fe^{3+} on the surface of SiO_2 , 3) heterogeneous reaction between crystallized iron oxide and H_2O_2 and, 4) reductive dissolution of iron oxide [1].

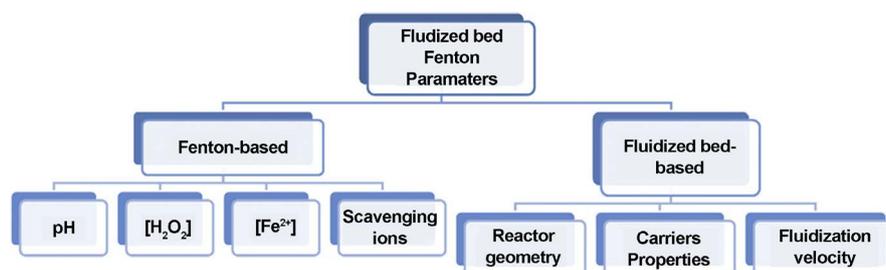


Figure 2. Main running factors of fluidized-bed Fenton process [1].

reagent to push the treatment method. Various choices may be acquired if electrochemical technology is implemented to Fenton oxidation. Between such, two manners could give an *in situ* production of Fenton's reagent. During the first manner, the electro-Fenton process, H_2O_2 is constantly formed at the cathode while the iron catalyst is externally introduced. The second procedure, the anodic Fenton process, implicates the utilization of sacrificial iron anode for the electrogeneration of Fe^{2+} while H_2O_2 is formed at the cathode or externally injected [1]. As numerous researches have usually grouped the latter as an electro-Fenton technology, it is really a peroxi-coagulation method, in which the contaminant is eliminated by the integrated oxidation and coagulation caused by the existence of $\text{Fe}(\text{OH})_3$ [79]. A thorough analysis of the electro-Fenton process and similar electrochemical techniques was published by Brillas *et al.* [79]; however, more fresh discussions of the basic concepts and environmental utilization of the electro-Fenton process were suggested by Poza-Nogueiras *et al.* [80] and Ganiyu *et al.* [81].

Usually, electro-Fenton is planned to possess a constant electrogeneration of H_2O_2 at the cathode. In a regular process, air/ O_2 is constantly furnished to the cathode; at the same time, the production of H_2O_2 is caused by a two-electron reduction of O_2 in acidic medium as illustrated in Equation(7) [63]. The introduction of Fe^{2+} leads to the catalytic degradation of H_2O_2 to form $\cdot\text{OH}$ founded on Fenton's reaction. Moreover, there is a constant electroregeneration of Fe^{2+} at the cathode as defined by Equation (8) [1].



As Fenton's reagent is produced *in situ* employing electron, which is a clean species, many of the restrictions of the classical Fenton oxidation may be avoided. With the constant *in situ* electrogeneration of H_2O_2 , the prices related to direct external injection of H_2O_2 will be avoided. Contrasted to the price of the external addition of H_2O_2 , Huang and Chu [107] affirmed that up to 80% cost decrease may be attained with the *in situ* electrogeneration of H_2O_2 . This price diminution will even augment when the costs related to transportation and storage of H_2O_2 are factored in [1]. This constitutes one of the benefits of electro-Fenton process contrasted to the conventional Fenton oxidation. An additional benefit of electro-Fenton process is the constant formation of Fe^{2+} (Equation (8)), which could reduce the potential sludge production from the precipitation of Fe^{3+} . Further, the constant regeneration of Fe^{2+} will diminish the catalyst input and improve the formation of $\cdot\text{OH}$ [63].

The performance of electro-Fenton is a function of numerous factors like applied current, solution pH, sort of electrolyte, the quantity of catalyst and the initial level of target contaminant. The impacts of solution pH, catalyst level and initial level of contaminant on electro-Fenton are identical to the classical Fenton oxidation. As an illustration, the optimum pH for efficient electro-Fenton was mentioned to be in the interval of 2.5 - 3.5 [74]. Applied current is a basic factor that touches the efficiency of electro-Fenton. The electrogeneration of H_2O_2 and the constant regeneration of Fe^{2+} are a function of the applied current. As a result, the performance of electro-Fenton augments with the augmentation in the applied current. Nevertheless, elevating the applied current over specific end may add parasitic reactions into the technique, decreasing the contaminant decomposition performance. The optimum quantity of applied current is application-specific and could be evaluated by early trials [1].

Several investigations have proved the performance of electro-Fenton to produce *in situ* Fenton's reagent for following decomposition of organic contaminants. Aside from the benefit of price diminution, efficacious decompositions of pollutants have been noted. On the other hand, the energy demand stays a worry in the electro-Fenton process [1].

3.3. Bio-Electro-Fenton (Microbial Fuel Cell)

The employment of microbial fuel cells (MFCs) to produce H_2O_2 for Fenton

oxidation, the so-named bio-electro-Fenton, is rising as an encouraging technique to diminish the cost linked to H_2O_2 purchase and transportation [77] [78]. In MFCs including an anode and cathode compartments, microorganisms are utilized to transform the chemical energy stored in organic pollutants to produce electric current and chemicals [108] [109]. The electrons are formed at the anaerobic anode compartment comprising the organic substrate and transported to the cathode compartment via an external circuit [110]. At the cathode, the two-electron oxygen reduction conducts to the generation of H_2O_2 . With the formation of H_2O_2 , Fenton catalyst (Fe^{2+} , iron oxide) can be introduced into the setup for the catalytic degradation of the H_2O_2 to form $\cdot\text{OH}$ [1]. **Figure 3** illustrates a schematic of a typical MFC employed in Fenton process (bio-electro-Fenton process). The simultaneous formation of H_2O_2 and electricity production make bio-electro-Fenton an interesting technique for wastewater treatment. Information about the basics of bio-electro-Fenton method may be read in the reviews published by Li *et al.* [82] [83].

The first published investigation on the bio-electro-Fenton was the research of Zhu and Ni [111]. They studied the decomposition of p-nitrophenol with simultaneous electricity production. Since then, numerous researches have noted the efficient employment of MFC to produce H_2O_2 for Fenton oxidation with concomitant electricity production. Mai and Li [110] concentrated on the usage of bio-electro-Fenton for decomposing Orange II at neutral pH. The maximum H_2O_2 formed was 3.24 mg/L, leading to total decomposition of the dye following 14 h and simultaneous power production of 230 mW/m². In identical research, Feng *et al.* [112] attained an H_2O_2 formation of 2.86 mg/L, conducting to total mineralization of Orange II. As an alternative of the dual-chamber usually utilized in the bio-electro-Fenton, Zhu and Logan [113] proposed a single-chamber MFC setup for decomposing phenol. The technology reduced more than 75% of total organic carbon (TOC) from the phenol-containing solution. Even if the

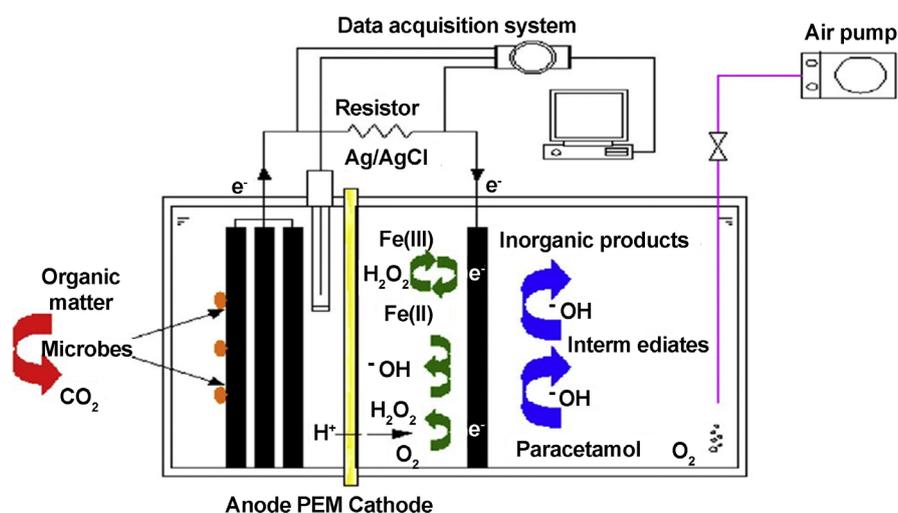


Figure 3. Schematic of microbial fuel cell applied to Fenton oxidation (PEM = Proton Exchange Membrane) [1].

single-chamber device is depicted to possess higher performance and lower operational cost, the quantity of H_2O_2 formed was not evaluated [1].

More fresh investigations of bio-electro-Fenton comprise the research of Li *et al.* [114]. Their main focus was the technique of employing a microbial reverse-electrodialysis electrolysis cell for decomposing Orange G. The fundamental characteristic of the device is that the electrons are formed from the exoelectrogens and salinity-gradient among seawater and freshwater in the cell. The technology conducted to the TOC elimination performance of 99.6%. Zhang *et al.* [115] tried the bio-electro-Fenton oxidation of paracetamol employing a dual-chamber MFC. The method eliminated until 70% of the initial paracetamol under optimum condition [1].

One main worry with the bio-electro-Fenton process remains the formation of O_2 as an alternative to the wanted H_2O_2 . To make sure the selectivity of cathode material for H_2O_2 generation, few scientists have proposed the employment of carbonaceous cathode as an alternative to the highly expensive Pt cathode. Carbon-founded materials are encouraging thanks to low price, plenty and electro-reduction activity. Wang *et al.* [77] [78] tried the reduction of bisphenol A, estrone, sulfamethazine and triclocarban employing bio-electro-Fenton with a graphite electrode. The maximum concentration of H_2O_2 formed was 2.06 mg/L. On the other hand, Asghar *et al.* [116] reached a maximum H_2O_2 production of 140 mg/L with a simultaneous electricity formation of 33.52 W/m² employing a heat-treated graphite cathode. Because of poor catalytic activity of carbonaceous material, higher yields of H_2O_2 has not been attained [1].

To obtain enhancement in the production of H_2O_2 , heteroatom doping of carbonaceous cathode is one of the proposed solutions [117]. In this attention, nitrogen doping has proved to be an efficacious procedure as it changes the electronic features of the carbon material and elevates the electrochemical surface area. Numerous researchers have established the contribution of nitrogen doping on H_2O_2 formation and power yields. For example, Asghar *et al.* [118] have proved a 25% augmentation in the yield of H_2O_2 formation through treating graphite cathode with NH_3 at elevated temperature. In addition, in many publications, H_2O_2 formation rates of 1.7 - 121 mmol/h/g are reached employing N-doped porous carbon [119]. Lately, Wang *et al.* [120] altered activated carbon air cathode by co-pyrolyzing with glucose and doping with nano-zero-valent iron to enhance two-electron oxygen reduction in the bio-electro-Fenton process. The changed electrode showed higher decomposition performance and more power production contrasted to pure activated carbon. Further, the anti-biofouling impact was noted on the altered cathode following an extended period of running [1].

Even if researches have proved the feasibility of the in-situ formation of H_2O_2 in the bio-electro-Fenton setup, there are numerous dares like the low generation of H_2O_2 , membrane fouling [121] [122], high internal resistance and reactor arrangements that require more revision prior its commercial implementations [1].

4. Electro-Fenton Process for Disinfecting Water

Ren *et al.* [123] tested a breakthrough by one method of new flow-through electro-Fenton with a graphene-modified cathode, which is frequently considered to be inefficient. They established that this technique was cost-effective for concomitant sulfadiazines (SDZs) decomposition and disinfection [124] [125] from urban secondary effluent with an extremely low electrical energy consumption (EEC) of 0.21 kWh/m³, explained to the elevated H₂O₂ formation of 4.41 mg/h/cm² on the fresh graphite felt cathode altered by electrochemically exfoliated graphene (EEGr) with a low EEC of 3.08 kWh/(kg H₂O₂). Contrasted with the inefficient SDZs decomposition by the traditional flow electro-Fenton, this technology was more cost-efficient and overpass the harsh needs of the electrolyte level. Further, it depicted excellent performance in decomposing diverse antibiotics, and the graphene-modified cathode still kept stable performance following eight subsequent trials. Taking into consideration the integrated work of [•]OH and active chlorine [126] [127] [128] [129] [130], the generation of hydroxylated and chlorine-containing by-products [131] was proved, and a likely decomposition route for SDZs was suggested. This flow-through electro-Fenton technique presented a substitutional process for killing pathogens and decomposing antibiotics through single technology for treating and reusing domestic secondary effluent (Figure 4).

5. Bio-Electro-Fenton Processes for Disinfecting Water

As mentioned previously, the bio-electrochemical technology-assisted advanced oxidation reactions (that is the bio-electro-Fenton system) have found an ideal position where they may be controlling in the next years, particularly for reducing recalcitrant organic contaminants. Contrasted to the classical electro-Fenton techniques, the bio-electro-Fenton setup hugely diminished the expenditures on wastewater treatment in terms of electric energy consumption and running prices. The bio-electro-Fenton setup is beginning to be a flexible technique providing a novel solution for emerging environmental problems linked with wastewater treatment. Li *et al.* [83] deeply examined the present publications concerning the decomposition of the recalcitrant organic contaminants in the bio-electro-Fenton device, particularly focusing on the treatment efficiency related to reactor design and major working factors.

As bio-electro-Fenton has been widely examined for eliminating recalcitrant organics, its implementation capacity vis-a-vis disinfecting water (that is killing pathogens) stays not familiar. Zhou *et al.* [132] tried the demobilization of *Escherichia coli* [133]-[139] in a microbial electrolysis cell-based bio-electro-Fenton setup (recalled as a microbial electrolytic-Fenton cell) with the objective to widen the utilization of microbial electrochemistry. They observed that a 4-log lowering of *E. coli* (10⁷ to hundreds CFU/mL) was attained with an external applied voltage of 0.2 V, 0.3 mM Fe²⁺ and cathodic pH of 3.0 (Figure 5). Nevertheless, unimportant demobilization was detected in the control tests

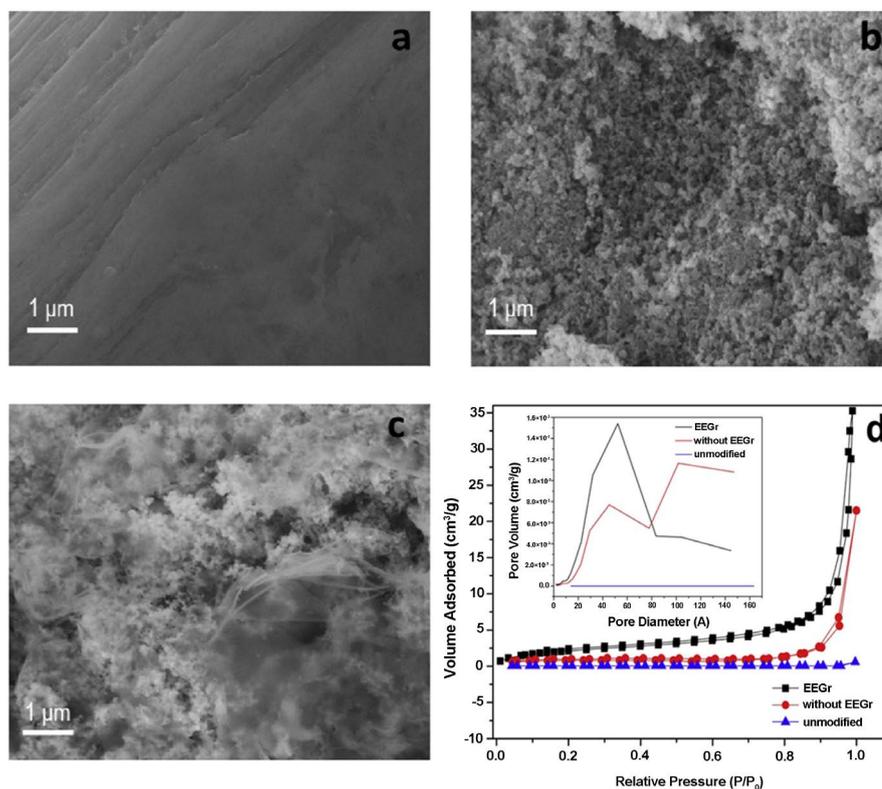


Figure 4. The Scanning electron microscopy (SEM) of unmodified (a), without electrochemically exfoliated grapheme (EEGr); (b) and EEGr; (c) modified graphite felts; N_2 adsorption/desorption isotherms and the pore size distribution of three different graphite felts (unmodified, without EEGr and EEGr) (d) [123].

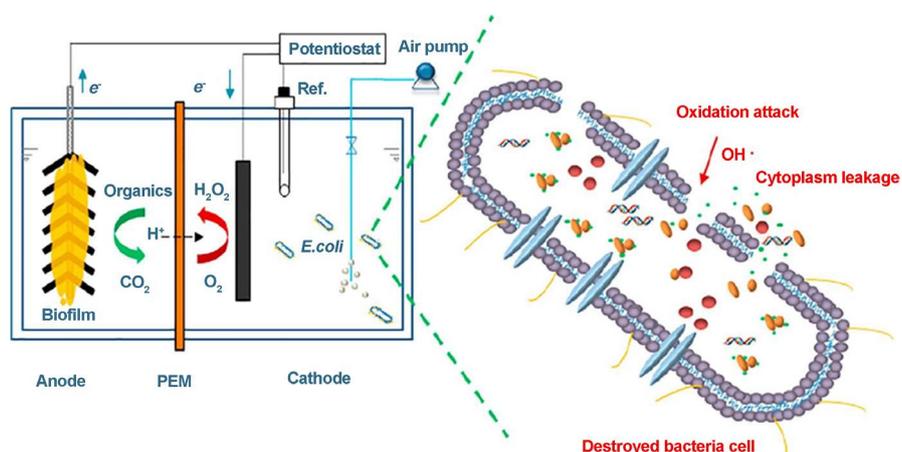


Figure 5. Schematic illustration of disinfection in Bio-electro-Fenton System [132].

without external voltage or Fe^{2+} injection. The killing microorganisms' impact was improved when the cathode airflow rate augmented from 7 to 41 mL/min and was also proportional to the elevation of Fe^{2+} level from 0.15 to 0.45 mmol/mL. Lethal cell film demolition via $\cdot OH$ was described as one probable route for disinfection (Figure 6). Zhou *et al.* [132] successfully established the likelihood of the bio-electro-Fenton technique for killing pathogens inactivation that provides

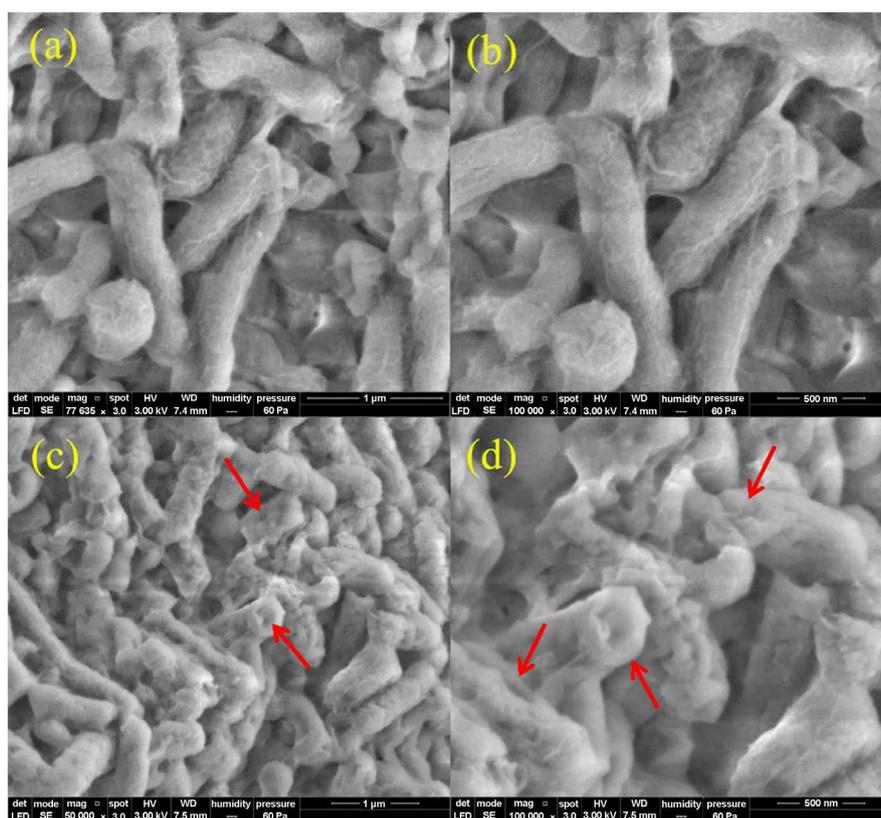


Figure 6. SEM images of *E. coli* (a) (b) untreated, (c) (d) after bio-electro-Fenton treatment for 60 min (arrows indicated that the collapses in cellular structure) [132].

comprehension for the coming expansion of possible, performant, and cost-effective biological [140] [141] water treatment technology.

6. Fenton Technologies: Hybridization as Main Future Trend

Despite the progress registered in the Fenton technologies, the classical process, and its ameliorated versions, adsorption and/or membranes processes [142] [143] [144] [145] remain fundamental for secure wastewater treatment.

In this context, Rogers *et al.* [146] established the chance of a toilet setup that recycles blackwater for onsite reuse as flush water, in which the blackwater is electrochemically treated to eliminate pathogens due to fecal pollution. Nevertheless, they found that this electrochemical technique needs great energy (48 - 93 kJ/L) to obtain total disinfection of the process liquid, and the disinfected liquid retains color and chemical oxygen demand (COD). Granular activated carbon (GAC) efficaciously diminishes COD in concentrated wastewaters. Rogers *et al.* [146] supposed that decreasing COD with GAC before the electrochemical application would enhance disinfection energy. They expanded and tried a hybrid system that integrates these techniques and proved its capacity to attain complete disinfection with elevated energy efficiency and liquid quality more appropriate for onsite reuse and/or discharge. Their hybrid setup combining both the pre- and post-treatment GAC filters with electrochemical treatment conducted

to a considerable decrease in the steady-state levels of numerous pollutants contrasted to the identical device without GAC (**Figure 7**). Most importantly, 1) the COD of the process liquid was diminished by 69%, and 2) the appearance of the water was hugely enhanced proved by the decreases in turbidity and color. The energy needed for total disinfection was diminished by 71% to 20 kJ/L with the addition of both pre- and post-treatment GAC filters. This energy decrease is probably attributed to the diminution in specifically soluble COD [146].

Lei *et al.* [147] suggested an electrochemical dynamic membrane filtration (EDMF) device for concomitant solid-liquid separation (also preserving electrodes against fouling) and sewage disinfection (**Figure 8**). At a low voltage of 2.5 V, efficacious disinfection was obtained in the EDMF, with ~100% log elimination performance (no detectable bacteria in the effluent). They proved that the EDMF setup, run at membrane flux of 100 L/(m²·h), may keep long-lasting bacterial disinfection performance of real wastewater (~100% log removal) in continuous flow trials (**Figure 9**). Transmembrane pressure (TMP) augmented from 0.8 kPa to 22 kPa during 80 d (one operation cycle), and cleaning of EDMF could efficiently reestablish TMP and biocidal actions for next filtration cycles. On the other hand, without dynamic membrane, the disinfection performance was reduced from the initial ~100% log elimination (with no detectable live bacteria) to ~44.4% log decrease during 7 d. Reactive oxygen species (ROS)-mediated oxidation was responsible for killing microorganisms in the EDMF, and [•]OH and H₂O₂ formed in this device had a key contribution, occasioning deterioration to cell envelopes and K⁺ escape from the cytosol. Further, catalase and superoxide dismutase for intracellular ROS attenuation was inhibited, leading to the augmentation of intracellular oxidative stress and therefore high-efficient disinfection. Such findings underline the capacity of EDMF configuration to be

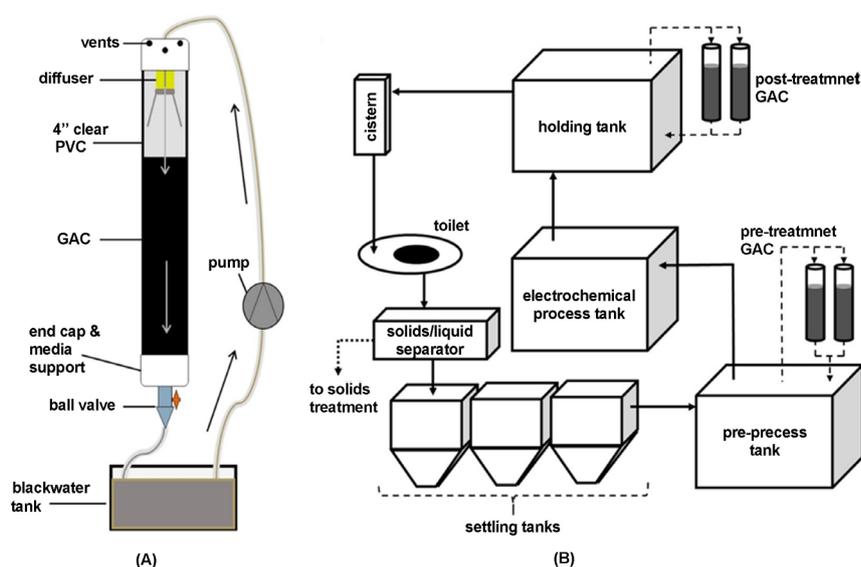


Figure 7. Experimental systems. (A) GAC packed bed column filter lab setup. (B) Schematic of GAC packed bed column filters integrated into the liquid treatment system [146].

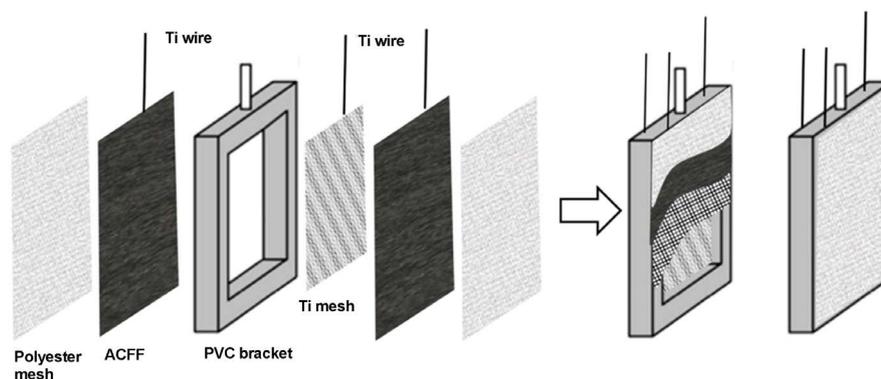


Figure 8. Schematic of the prepared EDMF module [147].

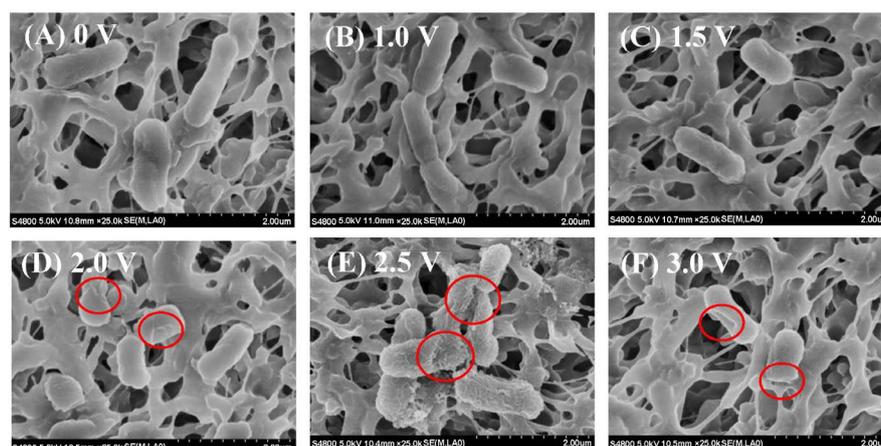


Figure 9. SEM images of *E. coli* after electrochemical treatment [147].

applied for treating and disinfecting wastewater.

Anfruns-Estrada *et al.* [148] suggested a sequential electrocoagulation (EC)/EF treatment (30 min each) as a more performant for a merged depollution and disinfection of urban wastewater.

7. Conclusions

From this work, the following conclusions can be drawn:

1) Restrictions like the need for acidic circumstance, sludge formation, and elevated chemical inputs have continued to retard the implementations of homogeneous Fenton in dealing with recalcitrant wastewater. Resolving such drawbacks stays, consequently, one of the significant research subjects in the domain of advanced oxidation processes for treating wastewater. This is obvious in the augmenting number of reports on procedures like heterogeneous Fenton oxidation, bio-electro-Fenton, fluidized-bed Fenton process, and homogeneous Fenton at circumneutral pH employing chelating chemicals. Between such procedures, heterogeneous Fenton remains the most largely tested thanks to the development in catalysis and material science. Fluidized-bed Fenton possesses the capacity to diminish sludge production and improve technique efficiency. A homogeneous Fenton process may be realized at circumneutral pH employing

chelating chemicals. Nevertheless, because of numerous problems related to the injection of chelating products, homogeneous Fenton at circumneutral pH has not attracted as much interest. Electro-Fenton and bio-electro-Fenton techniques have the potential to diminish the price linked with the external introduction of H_2O_2 and decrease sludge formation. Heterogeneous Fenton and fluidized-bed Fenton methods seem to be the most realizable strategies to deal with the drawbacks of the Fenton reaction. On the other hand, features like price, ecological consequences, and upscaling possibility will decide their large-scale usability. This work has briefly discussed the fresh progress in dealing with the restrictions of the Fenton process. It will, consequently, besides the excellent review of Bello *et al.* [1], serve as a reference document in the domain of Fenton reaction for recalcitrant wastewater treatment, especially for specialists looking for improving the effectiveness of Fenton response.

2) The microbial electrolytic-Fenton cell was established as an encouraging substitutional option for disinfecting and treating water [132]. H_2O_2 was *in situ* formed in the cathode and reacted with Fe^{2+} chemicals to excite the Fenton response. Device efficiency was touched by the applied voltage, cathodic aeration rate and the levels of Fe^{2+} injection. Batch trials divulged that the applied voltage of 0.2 V was seen as more appropriate for total demobilization. Further, disinfection impacts were proportional to Fe^{2+} portion and aeration rate; however, following economic considerations, 0.3 mmol (Fe^{2+})/mL and 29.8 mL/min aeration were adopted as optimal. As a prototype pathogenic microorganisms, *E. coli* hugely demobilized via the Fenton technology during which the bacteria cells were badly damaged through hydroxyl radical's aggression.

3) Despite the progress registered in the Fenton technologies, the classical process, and its ameliorated versions, membranes processes remain fundamental for secure wastewater treatment. As sure barriers towards pollution dispersal, processes such as nanofiltration should be coupled to Fenton techniques.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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