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Review

# Fenton: A Systematic Review of Its Application in Wastewater Treatment

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**Abstract:** The use of new technologies for the removal of pollutants from wastewater has become globally necessary due to the complexity and facilities defined by conventional treatments. Advanced oxidative processes, specifically the Fenton process, have become widely applied given their low cost and ease of use. Therefore, this study aimed to evaluate the progression of the scientific publications on the implementation of Fenton process, investigating their space–time evolution. Additionally, useful solutions, trends, and gaps in the applications for the removal of pollutants with this methodology were identified, and also different remediation strategies and the design of new treatments for wastewaters were identified within this scientometric analysis. Bibliometric research was conducted in two scientific databases, Web of Science and Scopus, from 2011 to 2022, and we identified 932 and 1263 studies with the word "Fenton," respectively. When these publications are associated with the treatment of alternative effluents, an increase in publications from 2011 ($r = 0.95$, $p < 0.001$) and 2013 ($r = 0.93$, $p < 0.001$) was observed when analyzing both databases, indicating the relevance of the theme. Among these studies, several of them were conducted on the bench scale (89.8% and 98.3%, Web of Science and Scopus, respectively) and in aqueous matrix (97.8% and 98.4%, Web of Science and Scopus, respectively), with being China the main country with publications associated with these words (28.33% and 41.9%), while Brazil is related to 3.65% and 2.29% of the total studies in Web of Science and Scopus, respectively. In addition, this review provides a guideline for new applications for different species in the matrices and describes the evolution of technological solutions to meet Sustainable Development Goal 6: clean water and sanitation.

**Keywords:** scientometrics; wastewater; advanced oxidative processes; pollution



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## 1. Introduction

According to the 2017 UNESCO report on water-resource development [1] contemporary production and lifestyles are closely associated with strict demands for high drinking water quality, but production and lifestyle have led to serious water pollution. Therefore, the same report states that about 80% of the wastewater is still globally returned to the environment without adequate treatment. Then, to mitigate the resulting pollution, regulatory agencies currently require on-site treatment of generated effluents. Several technologies are used for effluent treatment, including filtration, reverse osmosis, adsorption, and coagulation–flocculation [2–5]. However, new processes are being established to compare the results and increase the efficiency of the removal of persistent, refractory, and nonbiodegradable pollutants, such as drugs, pesticides, dyes, organic solvents, phenols, oily effluents, and agroindustrial wastes, such as stillage [6,7].

To degrade certain types of pollutants, chemical treatment, which generates oxidizing agents, particularly hydroxyl radicals ($^{\bullet}OH$), must be used. Currently, the most effective methods for degrading organic pollutants in wastewater and contaminated soil are advanced oxidative processes (AOPs), which include the Fenton approach. The Fenton process produces $^{\bullet}OH$ through a homogeneous reaction (takes place via the reaction from a mixture of hydrogen peroxide and ferrous salts). These oxidants are highly reactive and nonselective, reacting with a wide range of organic compounds and, consequently, degrading them into water, carbon dioxide, and inorganic ions [8]. For the by-products, the type of reaction that occurs between $^{\bullet}OH$ and the intermediate products will depend on the structure of the target compound.

Treatment processes based on the Fenton reaction have been effectively applied to treat various types of industrial effluents and municipal sewage treatment plants, as a single-, pre- and/or post-treatment methods [9]. For example, it can be used to treat leachate from landfill, from vinasse, as post-treatment of biodigested vinasse, in the treatment of effluents from the wood industry, in the removal of endocrine disruptors as well as to remove other environmental micro pollutants from the pharmaceutical industry, from hospital wastes, from university laboratories, from pesticides residues, and others [9,10].

The systematic literature review has become an established approach for comprehending the current state of knowledge on a particular topic [11–13]. This methodology enables not only the summary of the existing literature but also an assessment of spatial and temporal trends, as well as the identification of key biases and gaps, in the published research. It is important to note that some reviews do not bring details about the search methodologies to compose the review articles, which differs from the methodology adopted in the present work. For example, a general review on about the "Recent progress in Fenton/Fenton-like reactions for the removal of antibiotics in aqueous environments" was published in 2022 by Jiang and co-authors [14]. Nevertheless, the article outlines in general terms the applications of various Fenton oxidation technologies for the removal of antibiotics, the advantages, and disadvantages of these Fenton reactions in antibiotic removal. On the other hand, it described that Fenton-like oxidation requires less severe reaction conditions than Fenton oxidation, and it was more applicable to a wider range of processes, exhibiting lower economic costs and much higher cycle times. Additionally, it was emphasized that there is a simple and easy recycling method for the catalyst. Meanwhile, Wang and Zhuan [15] have also published the review of AOPs for the degradation of antibiotics without a specific bibliometric methodology. Conversely, the recent review follow the bibliometric methodology reported by other authors [16,17]. With this in mind, in this work a systematic literature review of the Fenton approach in wastewater treatment was carried out, with the aim of outlining its global use, reaction details, forms of data analysis, and temporal variations.

## 2. Methods

To identify relevant articles, a systematic literature review was conducted using the Web of Science (WoS, accessed on 3 January 2023) and SCOPUS databases (www.scopus.com, accessed on 23 January 2023). The search, in both databases, initially focused on papers with titles containing the terms Fenton* AND "industrial effluent*" OR wastewater* OR "water residual*" OR "persistent* compound*" OR "recalcitrant* compound*" OR "organic* load* " OR "chemistry oxygen* demand*" OR "vinasse" OR "hexaferrite nanoparticle*" OR "$Fe_2O_3$ nanoparticle *" until the year 2022. Only scientific articles were considered. Scopus is a comprehensive international database with unique advanced tools, such as ranking of journals, author profiles, number of articles published by a journal in a given year, and frequency of use of scientific terms.

The year of publication, journal, and country of the first author for each article containing the above terms were identified. The title, abstract, and keywords of the articles focusing on Fenton treatments were reviewed to obtain the following information: (i) the study scale (bench, pilot, or industrial); (ii) the use of Fenton alone or in combination with other treatments; (iii) the type of matrix (aqueous, soil, or air); (iv) reaction conditions (traditional, Fenton-like, nanoparticles, and various iron sources); (v) chemical oxygen demand (COD) and total organic carbon (TOC) removal percentages; (vi) compounds degraded by Fenton (dyes, pharmaceuticals, hygiene and cleaning products, phenol, pesticides, organic and inorganic compounds, vinasse, and other specific compounds); and (vii) the origin of the degraded effluents (industrial, hospital, sewage treatment plant, teaching or research institution, synthetic solution, or other specific origins).

To assess the change in the number of publications on Fenton over time, the Pearson correlation between the number of articles and the years that were published was determined. The article count was standardized by dividing the number of articles on Fenton published, each year, by the total number of articles in the WoS and Scopus databases for that year. We also used Pearson correlations to examine the trends over time in the types of effluents and compounds that Fenton has been able to degrade.

## 3. Results and Discussions

### 3.1. Time Trends in Studies Using Fenton

Considering WoS and Scopus databases, the search identified a total of 1073 and 1653 articles in indexed journals, respectively, until 2022, with the earliest indexed publication on the theme in Scopus dating back to 1992. Among these articles, 141 and 390 contained the term "Fenton" in their title but these did not employ the technique in their study. These papers were excluded from further analysis as they were not aligned with the objectives of this systematic review. In total, 932 and 1263 articles (WoS and Scopus databases, respectively) on the implementation of the Fenton technique to remove any form of pollutant were considered.

In regard to the use of the WoS database, it is important to indicate that WoS is mainly designed for researchers to find published literature, and it could be restricted for bibliometric studies. Then, it is not suitable to directly use all different types and levels of the WoS database for bibliometric study. For example, Data Citation Index, Derwent Innovations Index, Zoological Record, Social Sciences Citation Index, Arts & Humanities Citation Index, Conference Proceedings Citation Index—Social Sciences & Humanities, Book Citation Index—Social Sciences & Humanities, Current Chemical Reactions, and Index Chemicus. In fact, a minor number of articles was identified by the WoS search. Then, the information was collected once again with the same methodology previously used but the Scopus database was utilized.

Comparing the results obtained in the two databases (Figure 1), it is observed that WoS is less comprehensive than Scopus. This may be due to the Scopus database having a larger, internationally diverse database as well as a range of intelligent tools that can be used to aid the research process, such as journal rankings, author profiles, the number of articles published by a journal in a given year, and the frequency of use of scientific terms. Therefore, Scopus was used to elaborate the systematic review, examining the results obtained. Additionally, WoS results were reported as Supplementary Materials (SM) in order to illustrate the similarities.

Figure 1 illustrates the increase in the number of articles over time (Pearson's $r = 0.95$ and 0.93, for WoS and Scopus databases, respectively, with $p < 0.001$); a more pronounced growth in publications is attained since 2013 and, notably, in the years 2020 and 2022.

The findings revealed a trend over a period of thirty years in which the number of publications featuring AOPs has increased. This pattern is consistent with the macro-level explorations of chemical, electrochemical, photochemical, and photoelectrochemical technologies designed to remove organic pollutants from wastewaters [18]. The growing demand for the treatment of effluents that contain complex and recalcitrant pollutants is driving the use of the Fenton method [19]. Furthermore, the Fenton process is a versatile technique that can be combined with other technologies, as highlighted by Ramos et al. (2021) [20], who described the use of Fenton in a systematic review on textile dye treatment. Fenton is a homogeneous reaction, which occurs via a chemical reaction of the mixture of $Fe^{2+}$ and $H_2O_2$, which leads to the rise of homogeneous hydroxyl radicals ($^{\bullet}OH$) [9]. The use of Fenton is due to the growing demand for wastewater treatment containing increasingly complex and difficult to remove recalcitrant pollutants. It means that, with this type of oxidant, it is possible to treat different and varied ranges of residues, allowing for a low cost and reaction time, as well as avoiding the use of large technological devices [9].

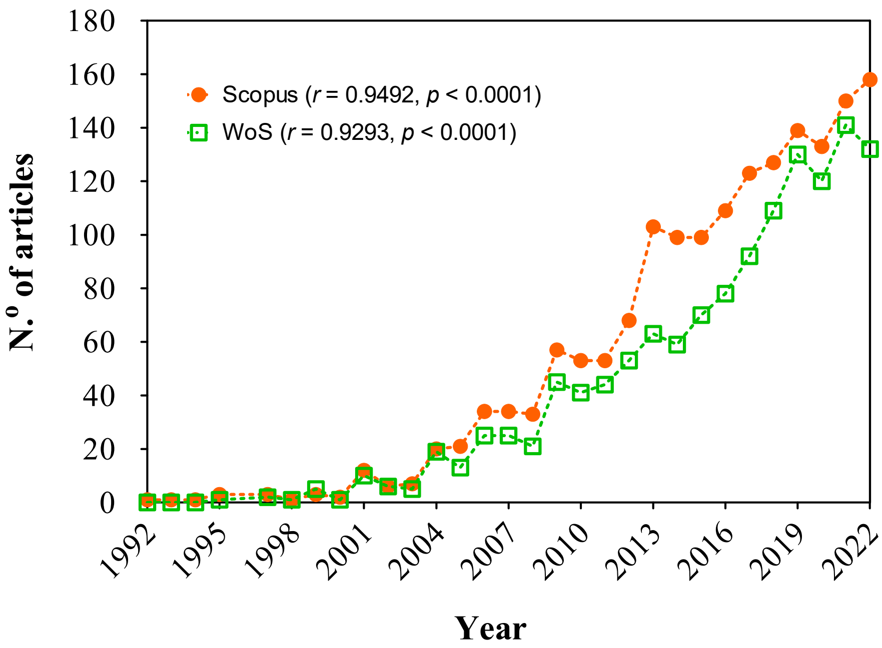


**Figure 1.** Comparison of the number of articles, between WoS and Scopus databases, using the same search methodology. Temporal trend in number of articles published in WoS and Scopus database.

From 2017 to 2022, researchers have incorporated statistical tools such as Takeuchi's method using the Mini Tap program, as well as the response surface methodology (RSM) with designs including the Box–Behnken design, Box–Wilson statistical design, central composite design (CCD), Central Composite Design in MINITAB®16 software, and other arrangements, such as the orthogonal array test and single-factor experiments, orthogonal experiments, and the regression quadratic equation [21–28]. Furthermore, the use of new reaction forms has increased, as described below.

Table 1 displays the nine categories of pollutants that were degraded using Fenton during 1992–2022, across 1263 studies. Among these studies, 90 focused on the use of Fenton to concurrently degrade various compounds, specifically those falling under the categories of pharmaceuticals, hygiene/cleaning products, and dyes.

**Table 1.** Temporal trend in number of articles published between 1992 and 2022, and Pearson correlation ($r$) in relation to number of articles published per year × type of compounds degraded by Fenton.

| Variable | No. of Articles | $r$ | $p$ |
|---|---|---|---|
| No. of articles | 1263 | 0.93 | <0.001 |
| Dye | 227 | 0.85 | <0.001 |
| Drugs | 114 | 0.81 | <0.001 |
| Phenol | 62 | 0.81 | <0.001 |

**Table 1.** *Cont.*

| Variable | No. of Articles | *r* | *p* |
|---|---|---|---|
| Organic compounds | 216 | 0.87 | <0.001 |
| Hygiene and cleaning | 11 | 0.42 | 0.02 |
| Pesticide | 22 | 0.59 | <0.001 |
| Inorganic compounds/metals | 40 | 0.82 | <0.001 |
| Vinasse | 5 | 0.32 | 0.09 |
| Other compounds | 418 | 0.69 | <0.001 |

The table above displays the results of studies that focused on the removal of pollutants using the Fenton technique. Among these studies, the majority focused on the removal of dyes (227; 18%), organic compounds (216; 17%), and pharmaceuticals (114; 9%); fewer were focused on removing phenol (62; 5%), inorganic compounds/metals (40; 3%), pesticides (22; 1.7%), hygiene and cleaning products (11; 0.87%), and vinasse (5; 0.39%). However, a large proportion of the studies (418; 33.1%) examined the use of the Fenton technique to remove a diverse range of other compounds, including solvents, polymers, organic molecules, wastewater from kitchens, dairies, slaughterhouses, tanneries, petroleum industry, mining, biodiesel, edible oil refineries, tertiary pulp treatment, wood industries and municipal effluents, bacteria, oilseed, and olive-oil industries [29,30]. We noted an increase in the number of articles over time for all compounds evaluated (Table 1); however, this increase occurred to a greater extent for dyes ($r = 0.85$; $p < 0.001$), organic compounds (0.87, $p < 0.001$), pharmaceuticals ($r = 0.81$; $p < 0.001$), phenol ($r = 0.81$; $p < 0.001$), inorganic compounds/metals ($r = 0.82$, $p < 0.001$), pesticides ($r = 0.59$; $p < 0.001$), hygiene and cleaning products ($r = 0.42$; $p = 0.02$), vinasse ($r = 0.32$; $p = 0.09$), and other types of specific composite products ($r = 0.69$; $p < 0.001$). These results are in agreement with the bibliometric analysis of WoS (see Figures S1 and S2 in the Supplementary Materials)

Regarding the treatment of wastewater with dyes, studies on this topic have increased in the last 10 years of the study period (2012–2022; $r = 0.85$ with time) [31–33]. Studies on the removal of organic compounds increased after 2015 ($r = 0.87$), showing that organic compounds were the most commonly used matrix in Fenton studies [34–36] in comparison with pharmaceuticals [37–39], phenols [40,41], pesticides [42,43], inorganic compounds [44,45], hygiene and cleaning products [46,47].

Figure 2 presents the sources of the pollutants listed in Table 1. However, 132 studies did not specify the origin of the treated matrix. Among those that were identified, 16 studies used the Fenton method to simultaneously degrade products from different sources, such as industrial effluent/laboratory-prepared solution (9), hospital effluent/laboratory-prepared solution (2), industrial effluent/other sources (1), industrial effluent/municipal wastewater treatment plant (WWTP) (1), and municipal WWTP/laboratory-prepared solution (2).

Most researchers have used Fenton for the degradation of industrial compounds (708; 56%), followed by laboratory-prepared solutions (synthetic effluent) (245; 19%), and other specific sources (98; 7.7%), such as sewage treatment plants (30; 2.4%), hospital sources (31; 2.5%), and research or educational institutions (3; 0.23%).

The volume of studies also correlates with the evolution and growth in the use of industrial processes and technologies over time, substantially increasing from the 2000s onward (Figure 1). Additionally, the search for new tools for sustainable industrial development involves introducing less environmentally harmful disposal techniques and then improving these techniques.

### *3.2. Size and Reaction Conditions of Fenton Reaction Studies*

Most studies on Fenton reactions have been conducted on the laboratory-bench scale, representing 98.3% of the studies analyzed, followed by those on the pilot and industrial scales. On the laboratory-bench scale, a smaller volume of effluent is used under controlled laboratory conditions, even though these effluents are representative of industrial effluents, as shown in Figure 2.

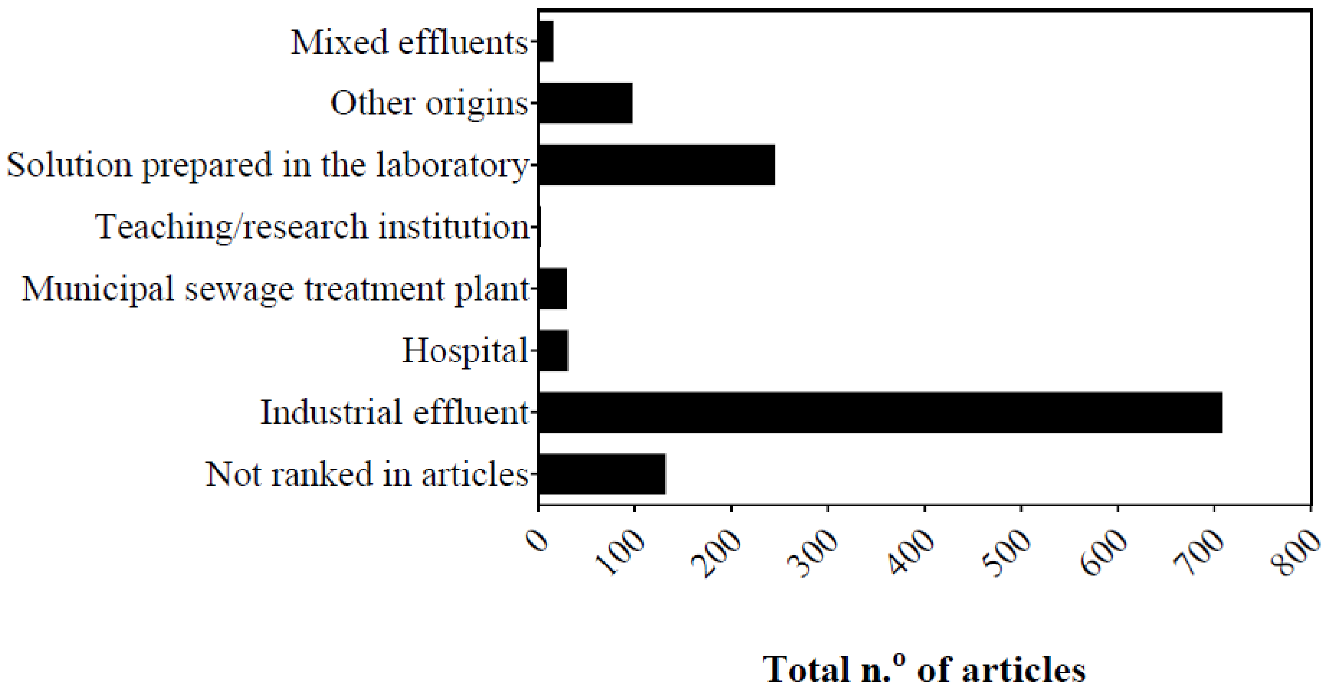


**Figure 2.** Temporal trend in number of articles published between 1992 and 2022 in Scopus database, considering origin of effluents degraded by Fenton.

Only 1.3% of the analyzed publications were conducted on the pilot scale, which is a method used for limiting the problems experienced with laboratory-bench-scale studies. However, pilot-scale studies are hindered by the operational costs associated with their implementation and execution. For example, Ferreira et al., (2020) [48] discussed the effect in a pilot plant process combining Fenton reaction with other AOPs and, then, the results showed that these kind of combinations are promising options for reducing the water footprint of biorecalcitrant industrial wastewaters. Moreover, 0.07% of direct studies have been conducted on the industrial scale, 0.07% of studies have used a combination of scales, and 0.31% of the studies did not specify the scale used.

Most studies (80.9%) used traditional reaction conditions (1022), followed by Fenton-like reactions (140) (Figure 3).

In the first case in Figure 3 on the y-axis, under ideal and optimized conditions, the compounds to be treated are catalytically oxidized in the presence of iron salts and hydrogen peroxide, in a pH range of 3.0 to 4.0. At values higher than 4.0, the rate of decomposition of hydrogen peroxide into water and oxygen markedly increases and the concentration of free iron species decreases due to the formation of iron complexes and precipitation of iron hydroxides. In the case of Fenton-like reactions, the catalyst can be other metals or complexes different from ferrous ion ($Fe^{2+}$). Various monovalent and divalent cations and complexes have been considered, as noted by Jonshon and Mehrvar (2022). We found that 0.87% of articles reported the use of $Fe^{2+}$ ions from secondary sources, such as industrial WWTP and mining sludge, whereas 1.3% of articles reported the use of Fe nanoparticles. The type of iron (hydro)oxide, whether natural or synthetic, solid, ionic, complex, or nano, affects the mechanism of $^{\bullet}OH$ formation. We identified this information in this review; according to Figure 3, the use of Fenton-like reactions with cation exchange, nanoparticles, and the combination of these conditions in the same treatment has been increasing. Pereira et al. (2012) [49] discussed catalytic forms of iron oxide [50], as well as other metal nanoparticles [51] and complex ions [52].

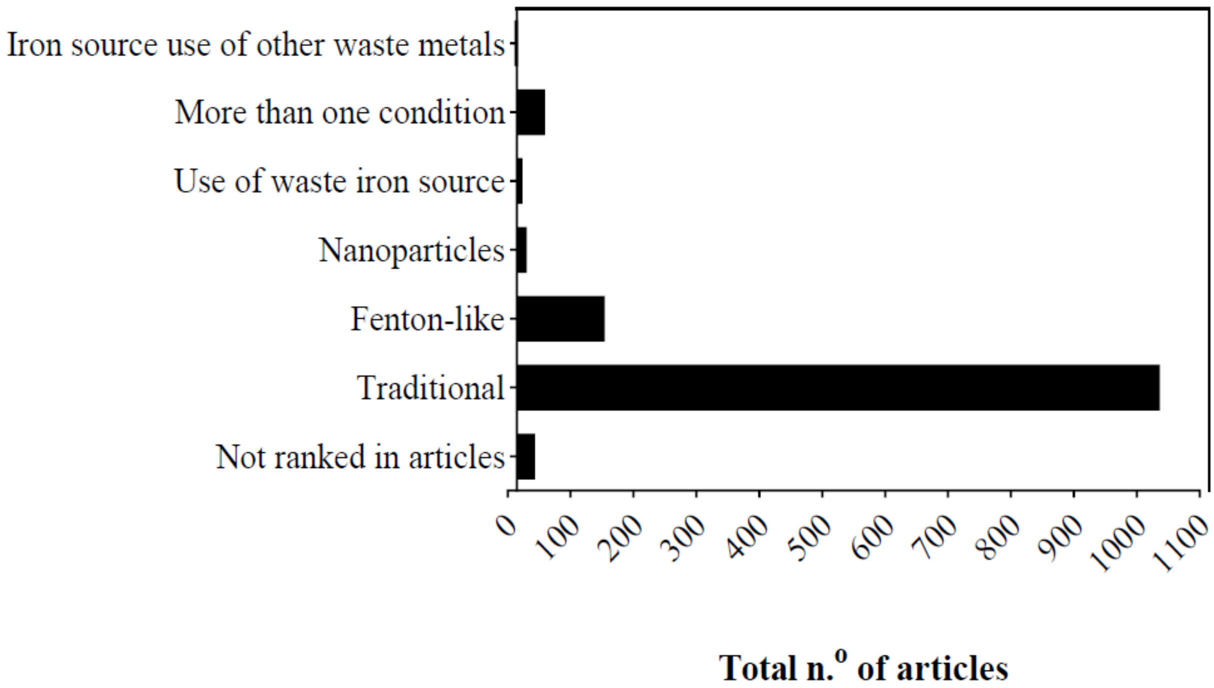


**Figure 3.** Fenton reaction arrangements on 30-year time scale.

For pH values below 3.0, the formation of hydroxyl radicals decreases, which becomes nonexistent at pH 1.0 [53]. Under these standard conditions, the reaction time is shorter and, consequently, the removal of compounds is lower compared with those of other AOPs [54]. Notably, only under acidic conditions does the $^{\bullet}OH$ radical dominate, which favors the chemical oxidation process. Moreover, under normal reaction conditions, a certain amount of flocculated material accumulates, which is subsequently removed by decantation, due to the formation and complexation of iron salts [55] and the formation of highly valent oxidizing species, such as $FeO^{3+}$ (Fe (V)) and the ferryl ion, $FeO^{2+}$ (Fe (IV)) [56]. The concentration of hydrogen peroxide in the reaction must also be evaluated, as the excessive use of hydrogen peroxide in the reaction medium acts as a scavenger of hydroxyl radicals. The concentration of iron ions is also important for the process, ranging from 3 to 15 mg/L as, under the above conditions, the scavenging of hydroxyl radicals occurs, which decreases the efficiency of the reaction [57].

In a single study, different reactive means can be combined with the aim of comparing their efficacy. In this survey, we found that in 3.5% of the cases, the efficiencies of the traditional Fenton reaction, Fenton-like reactions, and parallel-shaped nanoparticles were tested in comparison with that of a single process.

Regarding the amount of organic matter removed, in the majority of studies, 884, more than 50% of the COD and TOC, was removed. However, only 30 studies reported that the Fenton treatment removed less than 50% of the COD and/or TOC, which was also reported in the literature [58]. For 349 studies, the reduction in COD and/or TOC was not reported.

### *3.3. Isolated or Combined Processes with Fenton and Electro-Fenton*

Based on the articles analyzed, we collected information regarding the Fenton process being performed alone or in combination, accounting for 68.9% of the studies. Among these, 26.5% described the use of the Fenton process as the sole treatment [59,60], whereas 41.6% of the studies included Fenton combined with a second or third treatment [31,61]. Other techniques used for removing pollutants included electro-Fenton (EF) [10,62], photo-Fenton ($Fe^{2+}/UV/H_2O_2$) [63,64], post-biological treatment, pretreatment for subsequent physical–chemical treatment, ozonolysis ($UV/O_3$), photocatalysis ($TiO_2/UV/H_2O_2$), photolysis ($UV/H_2O_2$), catalysis, UV treatment, electrocoagulation, chemical coagulation, and membranes. The removal rate was increased when the Fenton process was combined with another chemical degradation technique [65].

Using the same search methodology, we observed that 20.9% of the studies focused on the EF process, with 13.6% of the total using EF as the sole treatment process [62,66,67]. Furthermore, studies on the combination of the EF process with other AOPs accounted for 6.9% of the total [59,68]. In contrast, the results for the use of the Fenton technique showed a different scenario. Fenton has been commonly used as the primary treatment process in various studies due to its reaction design, whereas EF is more restricted in its application, as it requires additional equipment such as reactors, electrodes, and power sources [18,62].

The fastest and most thorough destruction of target organic pollutants in water is currently achieved using Fenton-based electrochemical advanced oxidation methods [9]. The original method was EF; however, thanks to the occurrence of important photoreduction approaches, UV and solar photo-EF processes achieve a greater mineralization [9,53,62]. In actuality, the two-electron oxygen reduction reaction taking place at the cathode becomes ineffective or insufficient as solution pH rises, limiting the decontamination's efficacy [9,53,62]. Because of this, recent research has been concentrated on two key aspects of the EF processes: (i) developments in cathodic $H_2O_2$ electrogeneration, which demonstrate how the oxygen reduction reaction improves upon use of new and/or more sustainable electrocatalysts, cathode configurations, and reactor designs; and (ii) improvements in iron-based catalysts, with the primary goal of extending the application to a much wider pH range and ultimately surpassing the classical acidic limitation [9,53,62,69].

The electrochemical production of $H_2O_2$ is the basis of the Fenton-based EAOPs. Although the cathodic approach using the two-electron oxygen reduction process (Equation (1)) is considerably more common, this can also be created anodically [9,10,53]

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

After then, $H_2O_2$ can be either dropped in the medium to be progressively degraded, transferred to the anode to be oxidized to $O_2$ via the hydroperoxyl radical ($HO_2^{\bullet}$), or reduced cathodically to water [9,53,69]. The obtained cumulative $H_2O_2$ concentration depends on the cell configuration, the kind of cathode, and a number of operational factors. After that, $H_2O_2$ can be reduced cathodically to water, transported to the anode to be oxidized to $O_2$ by the hydroperoxyl radical ($HO_2^{\bullet}$), or left in the medium to gradually degrade. The cell configuration, the type of cathode, and several operational parameters all affect the cumulative $H_2O_2$ concentration that is achieved [9,53].

Despite the fact that $H_2O_2$ is a weak oxidant, the $Fe^{2+}$ ion promotes the occurrence of Fenton's reaction (Equation (2)). The greatest formation of free $^{\bullet}OH$ in the bulk is achieved at its ideal pH, which is close to 3. Because the resultant $Fe^{3+}$ may be continually reduced to $Fe^{2+}$ at the cathode surface via reaction (Equation (3)), continuing the catalytic $Fe^{3+}/Fe^{2+}$ cycle, this so-called EF process is more effective than the traditional Fenton [9,10,53,69].

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + {}^{\bullet}OH + OH^- \quad (2)$$

$$Fe^{3+} + e^- \rightarrow Fe^{2+} \quad (3)$$

PEF uses a UVA lamp to irradiate the solution as a photo-assisted method of EF. Reaction theory suggests that this increases the amount of $^{\bullet}OH$ produced as a result of the photolysis of $[Fe(OH)]^{2+}$, the main $Fe^{3+}$ species at pH ~ 3 (Equation (4)) [9,53]. Moreover, the UVA photons accelerate the mineralization of the contaminated solution by causing the rapid photodecomposition of the final Fe(III)-carboxylate complexes via the general reaction (Equation (5)).

$$[Fe(OH)]^{2+} + h\nu \rightarrow Fe^{2+} + {}^{\bullet}OH \quad (4)$$

$$[Fe(OOCR)]^{2+} + h\nu \rightarrow Fe^{2+} + CO_2 + R^{\bullet} \quad (5)$$

Instead, a UVC lamp can be employed, with the goal of producing more •OH through the photolytic homolysis of $H_2O_2$. Moreover, the aromatic contaminants may be photolyzed by the UVC irradiation, hastening their degradation [9,53]. Using the SPEF method, which uses free sunlight as the energy source, can fully eliminate the energy consumption associated with the use of expensive UV lamps, increasing the viability of the effective photo-assisted EF process. The substantially increased light power given by sunshine clearly outperforms the PEF method [9,53,69]. The experimental setups for these processes consist, essentially, of undivided two-electrode reactors with an air-diffusion cathode. For this reason, different oxidants produced at the anode can concurrently oxidize organic contaminants in an undivided cell [9,53]. While active chlorine ($Cl_2/HClO/ClO^-$) may also be formed by Cl oxidation in chloride medium, adsorbed M(•OH) is the most prevalent one in aqueous media. The primary oxidizing agents in anodic oxidation are M(•OH) and/or active chlorine, which may improve EF's capacity to degrade organics in the water matrix. The most relevant anodes for producing oxidants via the anodic approach are thought to be synthetic diamond electrodes [9,53]. Nevertheless, it is important to consider that the nature of the anodic material has a less significant role in the PEF and SPEF treatments, where the photons' action on the removal of the pollutants is more vital. Thus, PEF or SPEF pilot plants may employ substantially less expensive materials, such as dimensionally stable anodes [9,10,53,69]. Frequently, the total organic carbon decay and/or chemical oxygen demand of treated solutions indicate a significant efficacy of mineralization [9,53]. In this frame, the mineralization current efficiency, apparent current efficiency, and energy consumption demonstrate the treatment's feasibility. But, an increase in current accelerates the degradation and mineralization of the contaminants due to the quicker electrochemical synthesis of oxidants, although this effect is attended with a decrease in current efficiency and an increase in energy consumption due to the parasitic reactions [9,53,69].

It is possible to employ divided cells to create $H_2O_2$ in the catholyte, which is then kept for injection into a reactor where chemical Fenton treatment is then applied. Undivided cells are still the best choice to reduce cell voltage and save money on membranes [9,53,62]. For $H_2O_2$ electrogeneration in Fenton-based EAOPs via reaction (Equation (1)), there are traditionally two major cathode topologies to be distinguished [9,53]: (i) large surface area immersed/soaked porous structures or particle beds where the dissolved $O_2$ (from sparged $O_2$ or air) is reduced; and (ii) gas-diffusion electrodes (GDEs) formed by a gas-diffusion layer through which pumped atmospheric air or pure $O_2$ flows from an air chamber and a catalyst layer (i.e., a mixture of polymer with raw or modified carbon) where reaction (1) takes place [9,53,70]. When the gas encounters the solid and liquid phases at the three-phase boundary (TPB) [9,53,71], $H_2O_2$ is produced in both situations. The solubility of this gas restricts the yield when the former type is utilized, whereas the GDEs tend to be more effective and result in higher $H_2O_2$ concentrations due to the infinite supply of $O_2$ [9,72]. Other authors have merged the two designs to create a GDE that resembles three dimensions and is made of a hydrophobic carbon cloth on top of a porous graphite felt, achieving mineralization current efficiencies greater than 100% in SPEF [9,73]. Recently, a new version has been created that is theoretically more like the GDE [9,53,62]. It is made up of a floating (also known as air-breathing) cathode that is partially exposed to the environment to allow for natural air diffusion, negating the need for an air pump [53,74]. These cathodes are superhydrophobic (superaerophilic, in other words), and by increasing $O_2$ diffusion, they enhance the oxygen reductive reaction (ORR) catalytic performance [10,75]. By varying the polymer composition, air-breathing cathodes' hydrophilicity and hydrophobicity may also be adjusted, as can the catalyst's porosity. Zhang et al. (2020) [76] investigated the impact of cathode location on the generation of $H_2O_2$ revealing a higher current efficiency than a GDE. Recent work has focused on scaling up floating cathodes, which resulted in an $H_2O_2$ generation rate of 196.3 mg $h^{-1}$ [9,53,77]. It is important to note that there are alternative methods for also improving the feasibility of cathodic $H_2O_2$ electrogeneration for usage in EAOPs [9,10]. They are based on innovative reactor designs created for self-sustaining $H_2O_2$ generation, eliminating the need for the air pump [9,53]. Pérez et al. (2016) [78] have shown that a jet aerator based on the venturi (i.e., suction) action induced by a solution running at high velocity close to a tiny air intake exposed to the environment can produce enough $H_2O_2$. Scialdone et al. (2015) [79] and Hamdi et al. (2020) [80] suggested using pressured EF systems to supersaturate the solution with $O_2$ to decompose 8-hydroxyquinoline-5-sulfonic acid and Acid Orange 7 dye. One order of magnitude more $H_2O_2$ was produced during ORR at a graphite cathode using air compressed at 11 bar than at ambient pressure [9,79]. It has also been tried to combine the two reactor types mentioned, creating a pressurized jet-aerator. In another study, the same authors have also suggested a different reactor design to considerably boost the generation of $H_2O_2$ during the EF process. The $H_2O_2$ electrogeneration was improved by an order of magnitude using a microfluidic electrochemical device, which had an ideal nominal distance of 120 μm between the cathode (an inexpensive graphite plate) and the anode [81]. An alternative strategy is increasing the mass transfer of naturally dissolved oxygen to the cathode surface, for instance, using high turbulence promoters such as a revolving cylinder electrode as the cathode [82]. Another bright idea, tested with various anode–cathode configurations and liquid flow rates, attempts to use the anodically generated $O_2$ for further $H_2O_2$ synthesis in flow-through reactors [83,84].

The electrocatalysts typically demonstrate a trade-off between activity and selectivity, making it simple to make a decision that balances the amount of time needed to treat the water with the efficiency of the process (closely related to the $H_2O_2$ generation efficiency) [9,53,62]. It may be possible to simultaneously boost activity and selectivity for the formation of $H_2O_2$ by using heterogeneous single-atom catalysts with atomically scattered metal atoms on 2D materials [9,53,85]. An excellent illustration of this is a single Zn-centered phthalocyanine; however, the N ligands are gradually destroyed by $H_2O_2$, as is the case with porphyrins containing 3d transition metals.

Meanwhile, the kinetic current density of a Co-$N_4$ moiety inserted in N-doped graphene was increased by fine-tuning the atomic structure, with no activity loss for 110 h [86]. Yet, a significant shortcoming of single-atom catalysts continues to be their instability. Noble-metal-based electrocatalysts are also an excellent option for producing $H_2O_2$ because of their high activity, selectivity, and stability. There are also other options, such as Au-Pd [87] and Hg alloys with Pt, Ag, and Pd [88]. However, their complex manufacture, high expense, and usage of hazardous starting materials create a bottleneck for their actual use in water treatment. They may be ideal for producing $H_2O_2$ in small devices [9,53].

Carbonaceous cathodes are ideal for use in EF and related EAOPs [9,53]. Carbon catalysts are readily available, affordable, nontoxic, durable, and may be extremely reaction-selective (Equation (1)) [9,89] and the beneficial effects of surface imperfections, particularly the edges, have been experimentally demonstrated [9,10,53,90]. Although novel varieties of carbon-based and carbon-supported electrocatalysts have recently emerged, immersed carbon felt [9,53,91] and GDEs manufactured with carbon black (CB) particles combined with PTFE [9,10,53,92] are still the most widely used traditional carbonaceous cathodes for EF processes. As a strategy to boost activity, several scientists have adopted the switch from nanoparticles (like CB) to high-surface-area nanostructures. For instance, mesoporous carbons, CNTs, and graphene have all been employed in the EF process [9,53,93–95]. Macroporous (i.e., three-dimensional) carbonaceous cathodes are also widely used in order to support reaction (3), which would lead to continuous $Fe^{2+}$ electroregeneration [9,10,53]. Intentionally created fissures have also been used to create porosity [96]. Undoped and F-doped hierarchically porous carbon (HPC) have been recently created by Zhao et al. (2018) [97] as new structures with strong electrocatalytic activity to generate $H_2O_2$. These materials are created by the carbonization of metal–organic frameworks (MOFs), which produce an abundance of microporous, mesoporous, and macroporous materials rich in catalytic sites and improve the kinetics of ORR [98].

Several researchers have tried to modify carbons using various methods to improve their selectivity using organic solvents as well as chemical and electrochemical methods [99] and air calcination [100]. These modifications increase the cathode's hydrophilicity and electroactivity by causing oxygen and nitrogen functionalities [9,10,53]. For example, the electro-oxidation of graphite felt led to a sharp rise in $H_2O_2$ concentration from 3.2 mg $L^{-1}$ with the raw felt to 37.5 mg $L^{-1}$ at 90 min [99]. Meanwhile, the S doping of mesoporous carbon achieved a selectivity for $H_2O_2$ of about 80% [101], the P doping of carbon nanotubes [102], or the addition of B in addition to the traditional O and N [103]. Also, surface modification using quinones can be achieved, for example, with azo dyes, or Co-phthalocyanine [104]. The greatest concentration of $H_2O_2$, achieved at 100 mA $cm^{-2}$ (after 90 min of electrolysis in 0.1 M $K_2SO_4$ at pH 3.0), increased from 770.7 to 1025.9 mg $L^{-1}$ due to the inclusion of the azo dye [10,105]. Alternately, carbon can also become more selective when it is decorated with a variety of particle kinds. A number of chalcogenides have so far been explored, and co-structures are particularly well-suited for the generation of $H_2O_2$. Due to the great selectivity (80%) of $CoS_2$ with pyrite structure, GDEs built from decorated carbon nanotubes on carbon cloth or paper are now possible to produce [106]. Bronopol and bentazon can be degraded by EF and PEF since it has been demonstrated that the presence of phosphorus atoms in substitutional locations imparts a significantly stronger stability [107,108] as well as the particles made of W@Au [109] or NiFe produced from MOFs [110] have been employed. The recently discovered superhydrophobic cathodes are one example of how wettability/hydrophobicity is important. It can be possible by mixing silicon or a polymer with carbon to help trap more $O_2$ molecules, particularly in submerged cathodes. This effect has been demonstrated with the use of a breathable dimethyl silicon oil maturation layer, which lowers water flooding and increases rates of $O_2$ transfer, thus improving electrogeneration activity [10,111]. In contrast to cathodes that are exclusively hydrophobic or hydrophilic, a Janus cathode made up of a hydrophobic gas-diffusion layer and a hydrophilic catalyst layer was recently reported to increase the generation of $H_2O_2$, reducing the cost of the process [112].

It is important to note that one of the major trends in recent years has been the production of sustainable carbons, which has led to the development of greener electrocatalysts made from biomass [113]. According to the existing literature, these materials have mostly been examined for their ability to produce $H_2O_2$, utilizing bean deposits [114], glucose [115], or fungal hyphae [116]. Contrarily, Fenton-based EAOPs occasionally need cathodes, as was recently demonstrated for carbons generated from chitosan in the EF treatment of acebutolol [117].

Because acid pH is ideal for Fenton's reaction, the cathodes and reactors described in the preceding section have often been employed to treat polluted effluents via EF, PEF, and SPEF [118]. Yet, many industrial effluents and natural water have pH levels that are close to being neutral, which makes it difficult to apply traditional homogenous techniques. Two approaches may be used to overcome this disadvantage [119]: (i) heterogeneous catalysis using various types of solid iron, and (ii) homogeneous catalysis using soluble Fe(III) complexes (i.e., chelated Fe(III)). To create complexes with Fe, short-chain aliphatic carboxylic acids, like oxalic acid and citric acid, have been employed in the past. Due to their higher photoactivity, which enables a higher $Fe^{2+}$ regeneration, aminopolycarboxylic acids like ethylenediaminetetraacetic, nitrilotriacetic, and ethylenediamine-N,N′-disuccinic (EDDS) have recently been proposed as possible chelators. Due to its greater biodegradability, EDDS appears to be a preferable option among them. The continual electroreduction of the complex at a carbon–felt cathode surface, which is preferable than a GDE, has been credited with the high performance of the EF-like process using the Fe(III)-EDDS complex as catalyst [120]. The photoirradiation in a PEF system is an option to assure the production of Fe(II) [121]. All of these instances involve the progressive destruction of the organic chelator by $^{\bullet}OH$, even if the target pollutants can already be completely degraded before this happens [62]. It is possible to conduct heterogeneous Fenton-based EAOPs using natural minerals such as chalcopyrite and pyrite ($FeS_2$) [10,91,122,123]. For Fenton or EF treatments, crystalline pyrite is a particularly strong option since it is a superb electron donor whose ${S_2}^{2-}$ conversion to sulfate is followed by the release of $Fe^{2+}$ and $H^+$. Due to this, two degradation pathways—conventional Fenton's reaction, whose occurrence is encouraged by progressive acidification, and heterogeneous Fenton's reaction—can coexist. Nevertheless, considerable iron leaching restricts the pyrite-catalyzed EF's capacity to be reused and necessitates sludge management, despite the fact that it has demonstrated higher efficacy than other heterogeneous EF treatments [62]. In the case of vermiculite for dye treatment, iron-rich soils such as clays have also been researched [124]. Another source of solid iron is found in artificial iron-loaded materials, such as fly ash, modified cathodes, resins, zeolites, membranes, biosorbents, and hydrogels. Additional artificial structures include Fe-MOFs and Fe-MOF-derived carbons, which are potentially extremely porous nano/microcatalysts, and nanozerovalent ions [10,125–129].

A porous carbon modified with nanosized zerovalent iron (nano-ZVI) produced during pyrolysis of a 3D MOF was used. Its magnetic characteristics enabled recovery at the conclusion of the treatment, and it also demonstrated a high level of stability as shown by the little iron leaching and good recyclability. The utilization of an appropriately little amount (0.05 g $L^{-1}$) of a non-pyrolyzed 2D MOF in PEF treatment at pH 7.4 was granted. In this instance, adding carboxylic groups to the ligand improved the raw 2D MOF's resistance to hydrolysis [10,129].

### 3.4. Journals Publishing on Fenton Indexed in SCOPUS through 2022

The articles about Fenton were published in a total of 400 journals. The distribution of these publications was asymmetrical; Table 2 displays the 23 journals that had the highest number of publications in descending order, representing a total of 49.2% of the total publications.

**Table 2.** List of 23 journals with the most studies on Fenton until 2022.

| Journal | Total | % |
|---|---|---|
| Desalination and Water Treatment | 68 | 5.384 |
| Chinese Journal of Environmental Engineering | 67 | 5.305 |
| Journal of Hazardous Materials | 66 | 5.226 |
| Chemical Engineering Journal | 56 | 4.434 |
| Chemosphere | 38 | 3.009 |
| Journal of Environmental Chemical Engineering | 34 | 2.692 |
| Water Science and Technology | 34 | 2.692 |
| Environmental Science and Pollution Research | 27 | 2.138 |
| Journal of Environmental Management | 25 | 1.979 |
| Separation and Purification Technology | 25 | 1.979 |
| Water Research | 23 | 1.821 |
| Environmental Technology (United Kingdom) | 18 | 1.425 |
| Shenyang Jianzhu Daxue Xuebao (Ziran Kexue Ban)/Journal of Shenyang Jianzhu University (Natural Science) | 17 | 1.346 |
| Journal of Water Process Engineering | 16 | 1.267 |
| RSC Advances | 14 | 1.108 |
| Science of the Total Environment | 14 | 1.108 |
| Chung-kuo Tsao Chih/China Pulp and Paper | 13 | 1.029 |
| Journal of Cleaner Production | 13 | 1.029 |
| Journal of Chemical Technology and Biotechnology | 12 | 0.950 |
| Desalination | 11 | 0.871 |
| International Journal of Environmental Science and Technology | 11 | 0.871 |

**Table 2.** *Cont.*

| Journal | Total | % |
|---|---|---|
| Asian Journal of Chemistry | 10 | 0.792 |
| Processes | 10 | 0.792 |

To assess the quality of the articles and their publication venues, we conducted an evaluation using the Journal Citation Indicator (JCI). The JCI is a metric that is widely used to calculate the impact of a citation between articles and reviews published in a journal, considering the Journal Impact Factor, and reflecting the journal's performance over the past 3 years.

In this study, among the top 10 journals, four had a JCI above 1 (considered above average for their category): *Journal of Hazardous Materials* (1.94), *Chemosphere* (1.48), *Journal of Environmental Management* (1.38), and *Separation and Purification Technology* (1.43). *Chemical Engineering Journal* (2.13) stands out, with a JCI above 2, indicating that its performance was more than twice the average in its category.

In terms of the total number of publications (1263), the journals *Desalination and Water Treatment* (68), *Chinese Journal of Environmental Engineering* (67), and *Journal of Hazardous Materials* (66) were the most prominent, publishing a total of 15.91% (201) of the articles considered in the topic of this bibliometric analysis.

When considering the country of the corresponding author, the studies included in this review of Fenton research encompassed authors from 66 different countries. The distribution of publications among countries is uneven, with 10 countries accounting for approximately 75% of the total number of publications (as shown in Table 3). China produced the largest number of Fenton-related studies (530; 41.9%), followed by Turkey (95; 7.5%) and Iran (86; 6.8%). Meanwhile, Brazil ranked eighth with 29 published articles, representing 2.29% of the total.

**Table 3.** List of 10 countries that published the most studies on Fenton indexed in SCOPUS until 2022.

| Country | No. of Articles | Percentage |
|---|---|---|
| China | 530 | 41.96358 |
| Turkey | 95 | 7.52177 |
| Iran | 86 | 6.80918 |
| Spain | 62 | 4.90895 |
| India | 52 | 4.11718 |
| Portugal | 43 | 3.40459 |
| Taiwan | 38 | 3.00871 |
| Brazil | 29 | 2.29612 |
| Poland | 27 | 2.13777 |
| Italy | 18 | 1.42518 |

In terms of research and sustainable economic policies, China has established itself as a leader in the field, as reflected in its Scientific Electronic Library Online (2013) ranking. China is at the forefront of scientific-article production in the developing world, having experienced substantial growth in this area mainly owing to its exponential technological and industrial progress [130].

Additionally, several European countries, including Spain, Portugal, Poland, and Italy, are leading in industrial waste treatment within the bloc. This is due to the European Union's increased enforcement of environmental legislation, which has resulted in a higher number of scientific publications. Spain is a notable country on this list, with legislation based on the European Framework that covers the treatment of liquid and solid waste at all levels, strictly regulating waste and setting high standards [131]. Turkey has also achieved progress in industrial waste treatment owing to government support for projects focused on treating industrial effluents and recycling solid waste. Several Asian countries feature on the list of the top 10 countries publishing scientific research on waste treatment. Iran, India, and Taiwan can be noted for their strong regulatory environmental legislation [132]. In Brazil, which ranks eighth on the list, waste treatment has become a priority in recent years, and a consolidated legal framework is increasingly necessary to ensure compliance with environmental regulations and permits [133].

### 3.5. Perspectives on Water Reclamation after Fenton Treatment

The importance of water as a vital resource for humanity cannot be stressed at this difficult time of climate change. The annual demand for water is expected to increase because of the growing challenges that the world's population faces. According to estimates, by 2030, the global water demand would be 6900 billion $m^3$, or around 64% more than what most countries now have access to [134]. It is essential that the world prioritizes increasing the availability of clean water through alternative techniques, such as desalination, and water recycling and reuse. In the case of the treatment processes based on the Fenton reaction, these techniques have been effectively applied to treat various types of industrial effluents and municipal sewage-treatment plants in order to reduce the load of refractory pollutants. However, particular attention should be given to the side effects of the treatment, such as excesses of soluble ferrous ions.

Therefore, to ensure the availability and sustainable management of water and sanitation for all, water reuse and recycling as well as water reclamation after Fenton-based processes are essential strategies to accomplish SDG 6.

According to Domingues et al. (2022) [135], the total iron content accepted in Portuguese legislation is 2 mg $L^{-1}$. Therefore, several alternatives for the reduction of iron ions in the water matrices are being studied. For example, Volesky (2001) [136] has investigated the use of biomass, or Martins and co-workers (2017) [137] have studied the use of ion exchange resin. Both strategies can be steps in the Fenton post-treatment to suit the safety standards of potable water. It is worth mentioning that the possibility of Fenton-like reactions has been one of the most discussed reactions today [18], associated or not with the steps of reduction and adaptation to the proposed use of treated water [138]. In comparison to traditional Fenton procedures, the fluidized-bed Fenton (FBF) process offers a superior way for decreasing the creation of significant amounts of iron oxide sludge. It is frequently followed by the coagulation sedimentation (CS) process. In order to remove micro-pollutants and their by-products, FBF has found extensive use in the treatment of industrial wastewaters, including those from pharmaceutical, pesticide, textile, dye, and other refractory organic sources [139–141]. As a further low-cost and energy-efficient unit procedure, constructed wetlands (CW) have been suggested. According to Arden and Ma (2018) [142] and Liu et al. (2019) [143], CW systems integrate a wide variety of biotic and abiotic processes to offer a green solution for a variety of treatment applications. Before wastewater from wastewater treatment plants (WWTPs) is discharged into rivers, CW are frequently used to further treat the pollution [144]. This comprises effluents from many sources, such as home wastewater-treatment facilities [145], freshwater aquaculture facilities [146], the textile sector [147], and fertilizer-production facilities [148]. Previous investigations have demonstrated the effectiveness of CW in removing several pollutants and reducing important parameters, including chemical oxygen demand (COD, 78–81%), biological oxygen demand (BOD, 72–82%), total solid suspension (TSS, 79–89%), nitrogen (84%), color (74%), phosphorus (79%) [145–147], total organic carbon (TOC, 98.5%) [149], chlorpyrifos (94–98%) [150], organophosphorus pesticides (87.22 $\pm$ 16.61%) [143], steroid hormones (97.4 $\pm$ 0.09%), and biocides (92.4 $\pm$ 0.54%) [54]. Therefore, it is important to couple this strategy with Fenton-based technologies for real applications.

A typical urban river with little natural discharge (less than 5%) is a real example of this coupled strategy. The Shuangji River is found in the northern Chinese province of Henan. The greatest recycled paper manufacturing facilities in China are in the Shuangji River basin. Papermaking WWTPs dump approximately 75% of the river's recharge water, which severely pollutes the river's water system and causes the demise of aquatic species. A WWTP with CW was built with a daily capacity of 120,000 tons of wastewater treatment to address this problem. Then, FBF, CS and then the application of CW are all used in this integrated process, which has proven to be successful in lowering toxicity and raising water quality, eliminating the Fenton residues and, consequently, the water scarcity can be effectively addressed by using wastewater from the pulp and paper industries as reclaimed water. These conclusions were based on laboratory-scale and large-scale experiments, achieving under-optimal conditions for FBF treatment (pH (3.5), $H_2O_2$ dosage (0.93 mL/L), $H_2O_2/Fe^{2+}$ ratio (4), and hydraulic retention time (HRT, 60 min)), achieving average removal efficiencies of about 87.3%, 93.59%, 51.73%, 84.75%, and 95.86%, for COD, ammonia nitrogen ($NH_3$-N), total nitrogen (TN), total phosphorus (TP), and color, respectively. The amount of the genotoxic substance 4-nitroquinoline 1-oxide (4-NQO-EQ) was reduced from 30.6 ± 1.6 g/L in the influent to 12.4 ± 1.0 g/L after treatment by FBF, and then it was further reduced to 5.9 ± 0.4 g/L after treatment by CW and finally reached 3.2 ± 0.3 g/L after self-purification downstream for 12 km. Zebrafish chronic survival rates considerably increased over a 21-day period, rising from 0.0% in the influent to 58.8 ± 4.0% in the CW effluent before progressively increasing to 68.8 ± 2.6% after the 12 km downstream self-purification. Through this, the concentration of 8-hydroxy-2-deoxyguanosine in zebrafish dropped from 120.0 ± 19.3 ng/L in the ecological oxidation pond effluent to 94.0 ± 7.5 ng/L in the effluent of CW, and even further to 42.0 ± 3.0 ng/L after the 12 km downstream self-purification. In conclusion, the FBF-CW treatment method successfully detoxified wastewater from papermaking while also improving the quality of the water, making it appropriate for use as an ecological water supplement [151].

FBF techniques have gained popularity in recent years for the treatment of difficult industrial effluent. The advantages of sludge reduction provided by heterogeneous Fenton processes are combined with the efficiency of homogeneous Fenton reactions in FBF. FBF exhibits more promise in terms of cost-effectiveness and scalability when compared to other modified Fenton processes. However, the high Fenton reagent consumption rate and the requirement for exact pH control continue to be barriers to the full-scale application of FBF. Therefore, a classification for FBF processes has been developed into homogeneous, and heterogeneous approaches based on their response mechanisms and system designs as well as modeling methods like computational fluid dynamics models and artificial neural networks have been investigated because these have the potential to accelerate the full-scale adoption of FBF technology [152].

Another way to intensify the removal of organic matter is the combination of Fenton treatment with chemical coagulation techniques. For that, Metin and Cifci (2023) [153] reported that COD removal removed about 92%, and 96% of total suspended solids when a real effluent was treated applying this novel treatment strategy. In terms of cost, these authors determined that this combined treatment has a lower cost ($19.16 $m^{-3}$) when compared to treatment with photo-Fenton using UV irradiation.

The precipitated iron sludge can be affected by the environment of the reaction solution causing a negative impact on the practical application of Fenton technology. Iron catalyst deactivation, dosage amount additions and sludge-management issues [118] are also disadvantages on the use of the Fenton applicability. However, $Fe^{3+}$ is also a well-known flocculant, and some investigations have indicated that the hydrolysis of $Fe^{3+}$ also contributes to the removal of pollutants in the Fenton process. During the hydrolysis process, some pollutants can be removed by the coagulation and adsorption of $Fe^{3+}$ [154]. The homogeneous Fenton oxidation process followed by a pH neutralization to accelerate coagulation for the treatment of landfill leachate was utilized and 61% of COD was removed and the contribution ratio of Fenton oxidation and coagulation was 0.75 [154]. Meanwhile, a pesticide wastewater using the oxidation–coagulation process in the Fenton process was also treated, reducing the COD from 33,700 to 9300 mg $L^{-1}$, and, consequently, resulting in a significant improvement in biodegradability [154]. In addition to coagulation, iron salt can also play a role of adsorption to remove pollutants when these are hydrolyzed. The in situ-formed nano-hydrolyzed $Fe^{3+}$ particles (nano-FeOOH) have a higher specific surface area and can adsorb the carboxyl-rich intermediate product generated by Fenton oxidation [155]. The above results show that Fenton is a synergistic process of oxidation, coagulation, and adsorption. Normally, it is difficult to achieve the discharge standards of complex and refractory wastewater through a single treatment process. Then, multiple combined processes including adsorption, coagulation and advanced oxidation are often required.

Even though they are still made, direct olive mill effluents (OMW) are discharged into surface waters, which is dangerous, against the law, and has serious contamination effects. Thus, it is not acceptable to use OMW for irrigation [156,157]. According to several studies [158–161], the discharge of OMW has negative effects on the aquatic fauna and ecological balance as well as odor problems, soil contamination, plant-growth inhibition, underground leaks, water pollution, and disruption of self-purification processes [162]. For example, due to the significant amounts of pollution found in the Guadalquivir River Basin, the Spanish government outlawed the direct release of OMW into rivers in 1981. As a general remedy for OMW's natural evaporation, artificial lagoons were built in nations like Spain. The low evaporation rates, odor emissions, and dangerous leaks brought on by poor construction have, however, shown over time that this strategy is ineffective. Thus, chemical remediation techniques are required to clean up these resistant wastewaters [155–157,163–165]. In an olive oil mill in Jaén, Spain, which employs a cutting-edge two-phase olive-oil-extraction technology, earlier studies have improved a treatment process based on Fenton's reaction for the reclamation of OMW. The primary treatment (PT), which removes coarse particles, is followed by advanced oxidation with Fenton-like characteristics, flocculation–sedimentation, and filtration through olive stones in series (ST) [164,166–169]. However, the measured electroconductivity (EC) values in the effluent at the ST outlet (OMW/ST) are higher than the advised range of 2–3 mS $cm^{-1}$, indicating elevated salinity levels that pose a risk in accordance with the guidelines established by the Food and Agricultural Association (FAO) for using regenerated water for irrigation [170].

In recent years, membrane technology has also become a very attractive alternative to conventional separation methods, offering a number of advantages [171–173]. Although membrane technology has advanced significantly over the past 50 years, much work still needs to be done to improve its competitiveness in large-scale industrial applications. Membrane fouling is one of the key issues with membrane technologies, especially in wastewater-purification operations [162,174–178]. Fouling reduces flux quickly and jeopardizes the membrane's overall effectiveness. Due to the increased operational and energy expenses caused by this decrease in productivity, plants must frequently shut down for in situ membrane cleaning. Unfortunately, these cleaning techniques fall short of fully restoring the membrane module's initial effectiveness, leading to a gradual decline over time. Prior research on the reuse of OMW utilizing membrane technology has frequently noted problems such as concentration polarization and fouling formation on the membranes. These issues lead to a substantial loss of flux and permeate quality, which raises energy costs and makes the operation unprofitable. The selection of the wrong membrane types and insufficient operation conditions are the main causes of these fouling problems. In the study carried out by Ochando-Pulido and co-workers (2012) [162], the physicochemical properties of OMW after ST, which entails flocculation, filtering in series, and a Fenton-like reaction were investigated to determine the best membrane and avoid fouling during following pressure-driven membrane operations for the effluent's complete purification [162]. Analyzing bacterial growth, gauging saturation index, figuring out particle size distribution, and gauging the distribution of organic matter molecular weight cut-off (MWCO), the OMW ST was effective in reducing the dangerous electroconductivity (EC) values (2–3 mS $cm^{-1}$) and determining that the significant amount of organic contaminants (31.7%) with average diameters less than 3 kDa are relevant for membrane fouling. The saturation index suggested working with a recovery factor of less than 90%. Finally, using the chosen NF membrane at a pressure of 15 bar guaranteed low fouling, high flux generation (69.9 L $h^{-1}$ $m^{-2}$), and significant rejection efficiency (55.5% and 88.5% for EC and COD). This makes it possible to obtain an effluent of high quality in accordance with the FAO requirements in order to reuse the recycled water for irrigation.

Another study proposed a unique technology called advanced oxidation–nanofiltration hybrid technology in keeping with the developing circular economy concept. The method was created through experimental research on wastewater samples collected from a tannery factory in Kolkata [179]. A unique membrane made of graphene oxide (GO) is created especially for the nanofiltration stage, and the effectiveness of this membrane is contrasted with those of other commercially available membranes used to eliminate the residues after Fenton-based process. The reuse of the purified water was possible by subsequent microfiltration and subsequent nanofiltration. The synthetic GO-based membrane functioned remarkably well, achieving significant COD (99%), TDS (>96%), and chromium (>99%) removals. These findings demonstrate the promise of the novel GO-based membrane-equipped advanced oxidation–nanofiltration system as an efficient and sustainable method for treating wastewater from the leather industry, enabling its recycling and reuse [179].

To lessen the concentration of persistent pollutants, several industrial effluents and municipal sewage have been successfully treated using Fenton reaction-based processes. The presence of higher amounts of soluble ferrous ions, an issue in the wastewater treatment sector, is another one of the treatments' potential adverse effects that must be taken into account. According to studies by Domingues et al. (2022) [135], the highest allowable limit for the total iron concentration in wastewater released into natural water bodies or used as a source of public water in Portugal is 2.0 mg/L [135].

In a study by Victor-Ortega and co-workers [180], it was determined if Dowex Marathon C resin, a strong-acid cation resin, was effective at removing leftover iron from the supernatant liquid after iron precipitated during Fenton's oxidation of olive mill effluent (OMW). Their research concentrated on iron levels that were rather modest, with feed concentrations reaching up to 5 mg/L. According to the authors, the final effluent had an iron concentration of 200 g/L, which is less than the allowable limit for reusing water. In other related studies by the same authors, sodium, chloride, iron, and phenols were removed from a simulated OMW that underwent a chemical treatment involving Fenton's peroxidation and iron precipitation using Dowex Marathon C (strong-acid cation) resin and Amberlite IRA-67 (weak-base anion) resin.

A study on the efficacy of the Fenton reaction in treating wastewater from the paper industry found that the removal efficiencies for TSS, BOD, COD, TKN, and total phosphorus (TP) were all very high, with values of 95.4%, 92.7%, 93.7%, 90.5%, and 91.7%, respectively. This study showed Fenton treatment's effectiveness in permitting the reuse of paper industry wastewater for irrigation while meeting Egyptian code irrigation criteria [181]. The goal of the current study was to increase greywater treatment's effectiveness by using a chemical-treatment strategy. In order to decrease the physicochemical properties of the greywater to levels appropriate for irrigation, as determined by the applicable regulations, this chemical treatment involved the employment of Fenton's reaction as an advanced oxidation process.

According to Ribeiro et al. (2016) [182], clothes colored using wastewater treated with Fenton's reagent (reused water) produce results that are comparable to those of fabrics dyed using conventional dyeing techniques (production waters). When it came to the pink and gray fabric samples, Fenton's wastewater treatment produced even better results than reclaimed water, which, because of its natural hue, significantly darkened the tone of the materials. All fabric samples passed stringent industrial quality control, and the results were all completely satisfactory. The fabrics made utilizing wastewater treated with Fenton's reagent did not exhibit any instances of stains, discoloration, opacity, or any other unfavorable impacts. These results show that wastewater that has also undergone post-treatment is suitable for reuse in the textile-dying procedure.

The discussion above makes it very evident that waste is produced by all water purification and recycling methods, especially the Fenton process. As a result, waste management is crucial for both the environment and human health. This is due to the fact that waste materials are typically poisonous. The produced waste could contaminate water supplies once more. Screening, filtering, centrifugal separation, sedimentation, gravity separation, coagulation, flotation, aerobic and anaerobic processes, evaporation, and precipitation all produce a lot of solid waste. Water with a high concentration of contaminants is produced via reverse osmosis, micro-, and ultra-filtration, and Fenton-based processes. For the management of waste materials produced during water-treatment procedures, numerous strategies have been devised and used. Recycling sludge into useful items like fertilizers, fillers, building materials, etc., is the most crucial waste-management strategy. A few hazardous waste materials have been burned, and the ash from those fires has been used as fertilizer. It has also been suggested to bury the waste materials underground in containers constructed of hermetic plastic or iron. These days, beneficial and affordable adsorbents are utilized to remove contaminants from water, including fly ash, red mud, and sand, and these can be used after Fenton treatment. Large amounts of these exhausted adsorbents have been manufactured, and they have been successfully used as fillers and building materials.

## 4. Conclusions

The environmental impacts caused by the waste generated by industries worldwide require attention and specific treatment that is of low cost and easy to operate, and that combines the efficient removal of recalcitrant carbon and other micropollutants, both on medium and large scales. Through this survey, we identified a substantial increase in publications focusing on the Fenton method from the 2000s onward. However, most of these publications are still concentrated in China and Europe, highlighting the need for more incentives for the treatment of alternative waste in other countries.

As suggested in the literature, the Fenton process is an effective and cost-efficient method for treating a diverse range of compounds found in various types of effluent matrices. This process particularly efficient at removing COD, with satisfactory results of over 50% achieved in most studies. The results of this review showed that one of the most common applications of the Fenton process is in treating dyes and industrial effluents in general. Another feature that must be considered is that EF is a Fenton-based technology which has received great attention in the last decade, allowing significant technological advances on the reactors and catalysts [10].

This systematic survey also demonstrated the need to conduct studies on larger operational scales, so that the results are effective and applicable in industrial settings; most studies have been conducted on the laboratory scale, lacking adequate transposition to larger scales. Thus, this review can serve as a benchmark for the initiation of new studies involving the Fenton process, helping to establish it as an important chemical process for the treatment of effluents.

**Supplementary Materials:** The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/pr11082466/s1, Figure S1: Time trend for the number of articles published between 1995 and 2020 in the Web of Science database, considering compounds degraded by Fenton; Figure S2: Time trend for the number of articles published between 1995 and 2020 in the Web of Science database, considering the origin of effluents degraded by Fenton; Figure S3: Types of scale in which the Fenton studies were carried out; Figure S4: Types of matrices in which the Fenton studies were carried out.

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