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19

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A global trend of Fenton-based AOPs focused on wastewater treatment: a bibliometric and visualization analysis

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Abstract

This study sought to detect the global characteristics and developments of Fenton-based advanced oxidation processes (AOPs) focused on wastewater treatment. Using bibliometric and visualization analysis via VOSviwer, CiteSpace, and HistCite software, it investigated and analyzed different aspects, including growth trends, country, journal, category, and keyword analysis, as well as citation-burst detection of references to 5,940 records ranging from 1990 to 2019 in the Web of Science Core Collection. It was concluded that wastewaters in the textile industries, pharmaceutical industries, phenol compounds, and landfill leachates were currently and would continue to be the most focused industrial effluents that can be treated efficiently by applying Fenton-based AOPs. The study also conducted an in-depth analysis based on four clusters through the 100 most-occurring keywords, containing textile wastewater, catalyst, photo-Fenton, and electro-Fenton, identifying the four most studied research topics of Fenton-based AOPs. Studies about innovative (nano) catalysts, electro-catalytic, and photocatalytic materials can provide new degradation possibilities and overcome the Fenton process's drawbacks. The results of this bibliometric analysis can be helpful information for researchers and industry practitioners.

Key words: bibliometric analysis, citation burst, Fenton-based AOPs, visualization analysis, wastewater

Highlights

- Present comprehensive bibliometric information about Fenton-based AOPs for researchers and industry practitioners.
- Detect improvements on Fenton-based processes during 28 years.
- Using histcite, Citespace and VOSViewer for detecting global trends of Fenton-Detect improvements on Fenton-based processes during 28 years.
- Detect 20 most cited references on this area of research.

INTRODUCTION

Industrial wastewater containing hazardous compounds is increasingly introduced into the environment because of the continuous development of new products, technologies, and industrial processes. Such streams often contain biorefractory and toxic pollutants. Recently various new technologies have been developed to remove such damaging effects of these pollutants on the environment, and researchers continue to try to find ones at a lower cost. In wastewater treatment processes, except for biological and chemical ones, the pollutant only transforms into another phase, resulting in secondary loading on the environment (Saharan *et al.* 2014). On the other hand, biological processes are also not always practical because of the recalcitrant nature of some

Water Practice & Technology Vol 16 No 1
doi: 10.2166/wpt.2020.099

pollutants, and some of the organic compounds exhibit toxicity toward microorganisms (Eljarrat & Barcelo 2003). While direct chemical oxidation can effectively degrade biorefractory substances, the operational cost of this process is high (Levec & Pintar 2007). Among alternative treatment technologies, advanced oxidation processes (AOPs) present considerable potential for degrading several pollutants, and over the last two decades, AOPs have received significant attention for degrading a large number of organic pollutants, specially biorefractory compounds (Antonopoulou *et al.* 2014; Ribeiro *et al.* 2015). AOPs that consume less energy than direct oxidation (Levec & Pintar 2007), are based on the in-situ generation of highly reactive oxygen species (ROS) like the hydroxyl radicals (HO•), which have a standard redox potential of 2.8 V1 (Pignatello *et al.* 2006a). Because of the diversity of ROS, such as hydroxyl radicals, H_2O_2, O_3 and superoxide anion radicals ($O_2^{\bullet-}$), along with numerous means for ROS production, such as chemical, photochemical, and electrochemical processes, several different AOPs are available (Bernal-Martínez *et al.* 2010). According to several studies, especially (Kahoush *et al.* 2018; Khatri *et al.* 2018), AOPs can be categorized into Fenton-based, UV-based, O_3-based, and electro-oxidation and physical processes, as shown in Table 1. Most of these use a combination of potent oxidizing agents with catalysts (e.g. metal ions) and irradiation (e.g. UV, visible light) and are quite similar in using the presence of HO• to improve the treatment process (Antonopoulou *et al.* 2014). In most of these methods, the target pollutant can be mineralized entirely into CO_2, H_2O, and mineral acids (Antonopoulou *et al.* 2014; Ribeiro *et al.* 2015). Among these versatile AOPs, Fenton is most commonly used for the degradation of wastewater pollutants.

Table 1 | Classification of different AOPs

AOPS				
Fenton-based	**O_3-based**	**UV-based**	**Electro-oxidation**	**Physical**
Fenton Photo-Fenton Electro-Fenton Solar-photo-Fenton Photo-electro-Fenton Sono-electro-Fenton Sono-photo-Fenton Fenton-like or catalytic Fenton	$O_3 + H_2O_2$ O_3 + catalyst electro-peroxone*	UV + H_2O_2 UV + Cl_2 UV + O_3 UV + SO_4 UV+ catalyst	TiO_2-doped electrodes SnO_2-doped electrodes PbO_2-doped electrodes Boron doped diamond electrodes	Electron beam Ultrasound Plasma Micro wave Hydrodynamic Cavitation

*$O_3 + H_2O_2$.

The Fenton reagent (an aqueous combination of H_2O_2 and Fe^{2+}) was first discovered in 1894 by the chemical engineer Henry John Horstman Fenton (Fenton 1894).

As described in Equation (1), this reagent, in acid medium, leads to the decomposition of H_2O_2 into active oxygen species capable of oxidizing organic pollutants; for example, the oxidation of Fe^{2+} to Fe^{3+}(Bossmann *et al.* 1998).

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + OH^- + OH^{\cdot} \quad (1)$$

Fe^{3+} formed in Equation (1) can react with the H_2O_2 present and form Fe^{2+} again, producing OH•, as shown in Equation (2) (Walling & Weil 1974).

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

Water Practice & Technology Vol 16 No 1
doi: 10.2166/wpt.2020.099

This reaction occurs more slowly than reaction 1 (Pignatello 1992), and the Fe^{3+} ions also react with the $HO_2^{\cdot}$ and thereby reduced to Fe^{2+}, as shown in Equation (3).

$$Fe^{3+} + HO_2^{\cdot} \rightarrow Fe^{2+} + O_2 + H^+ \tag{3}$$

There are many advantages in using the Fenton process (Pignatello *et al.* 2006b; Barbusiński 2009; Telles & Granhen Tavares 2012; Kahoush *et al.* 2018):

(1) the reaction time is relatively short;
(2) it can be carried out at room temperature and atmospheric pressure;
(3) the Fenton reagent is readily available at reasonable prices, easy to store and handle and safe; and
(4) hydroxyl radicals are produced chemically without any energy input.

However, some disadvantages limit its use; for example, pH control is needed to keep the pH range around 3, approximately 50–80 ppm of ferrous ions is required, and sludge streams containing chemicals such as iron salts are continuously produced, with the process sometimes inhibited because iron ions can be consumed faster than their generation (Kahoush *et al.* 2018).

Because of these drawbacks, many different Fenton process modifications have been developed, in this study categorized as Fenton-based AOPs (Table 1). There is also a substantial body of published papers related to the application of Fenton-based AOPs in wastewater treatment, and this volume requires effective management.

Bibliometrics is a mathematical tool that is combined with statistical methods and can be used to enumerate and analyze publications in the context of a particular subject within given categories such as topic, field, source, institute, author, or country (Dai *et al.* 2015). While it is a powerful tool that assists in using keywords to assess developmental trends or future research directions (Han *et al.* 2014), it is unable to support in-depth analyses such as cluster analysis (Li *et al.* 2018). Information visualization analysis, on the other hand, is a process for representing raw data in a visual and meaningful way (Hou 2014), so finding more accurate and comprehensive results can be made possible by combining bibliometrics with information visualization. There have been many studies applying these two analysis tools that focus on environmental issues. Liu *et al.* investigated the bibliometric analysis of global biodiversity during 1900–2001 (Liu *et al.* 2011). Their study revealed the adoption of advanced technologies, and demonstrated interest in the patterns, as well as underlying processes of ecosystems. Malarvizhi *et al.* studied the research trend in adsorption technology (Malarvizhi *et al.* 2010). They indicated the global trend of dye removal over 16 years. Li *et al.* investigated climate change through bibliometric analysis (Li *et al.* 2011), and they showed significant directions of climate change in the 21st century containing temperature, greenhouse gas and precipitation. There are also some other researches on environmental issues such as water resources (Wang *et al.* 2011), wetlands (Zhang *et al.* 2010), solid waste (Fu *et al.* 2010), desalination (Tanaka & Ho 2011), and aerosol research (Xie *et al.* 2008), as well as energy (Li *et al.* 2017). Still, there are none on Fenton processes.

As mentioned, there is a large amount of publications on Fenton processes. There are many review papers on this area of research but there is no bibliometric research. In this manuscript, in-depth analysis of top keywords in Fenton-based processes was conducted. Therefore, improvements on Fenton-based processes during 30 years have been detected, and it has provided comprehensive bibliometric information about Fenton-based AOPs for researchers and industry practitioners. Since there was a need for multi-perspective study research on applying Fenton-based AOPs to wastewater treatment, this study applied a combination of bibliometric and information visualization analysis to detect characteristics and trends of this topic between 1990 and 2019.

Water Practice & Technology Vol 16 No 1
doi: 10.2166/wpt.2020.099

The objectives of the bibliometric analysis are:

- determination of year-wise trends and the country-wise distribution of research on the topic of concern and the collaboration between them as well as identifying the significant journals;
- categories and keywords analysis; and
- detecting references with the strongest citation bursts.

METHODS

To evaluate the historical trends of studies on Fenton-based AOPs applied to wastewater treatment, a bibliometric analysis was conducted using the Web of Science Core Collection over the time interval 1990 to 2019. The Web of Science Core Collection presents a complete and accurate view of over 115 years of the highest quality research in terms of author information, journals, and a citation for bibliometric analysis. (Fenton) and (wastewater* or waste-water*) were used as search phrases for searching topics in the Web of Science Core Collection from 1990 to 2019, and after finding all such publications, records in languages other than English were excluded, resulting in 5,940 records fully downloaded as a database for further analysis. Data analysis of publications, countries, and categories was performed using Excel and HistCite software (Garfield 2009), and visualization of collaborative networks among countries was conducted using VOSviewer software (van Eck & Waltman 2010a). To analyze the keywords, VOSviewer and Citespace software were applied to discover keyword clusters and citation bursts of references, respectively (Chen 2006; van Eck & Waltman 2010b).

RESULTS AND DISCUSSION

Time-wise and country-wise analysis of publications

In all, 5,940 works from 95 countries on wastewater treatment applying Fenton-based process were published from 1990 to 2019.

Time-wise trends of the top 10 most productive countries were analyzed with the results shown in Figure 1. It can be seen that, before 2007, Spain had been the most productive country, but then the People's Republic of China (PRC) surpassed Spain and remained in first place in terms of publication numbers. Despite some fluctuations, the publication numbers of countries kept increasing annually. To evaluate the historical trends of studies on Fenton-based AOPs applied to wastewater treatment, a bibliometric analysis was conducted using the Web of Science Core Collection over the time interval 1990 to 2019. The Web of Science Core Collection presents a complete and accurate view of over 115 years of the highest quality.

To evaluate the quality of publications, Table 2 listing the top 20 most productive countries was created, where the total local citation score (TLCS) and total global citation score (TGCS) were introduced as metrics. TLCS represents the total number of citations of all publications from a specific country based on local records. At the same time, TGCS shows the total number of citations of all publications appearing in the Web of Science from a specific country (Garfield 2009). Interestingly, although the number of publications of PRC was about twice that of Spain, the TLCS and TGCS of publications in Spain were the greatest. Generally, this figure reveals that for a specific country, there seems not to be a meaningful connection between citation and publication numbers.

The collaborative network between all the named countries is depicted in Figure 2, in which circles and curves are representative of countries and the cooperative relationships between two connected countries, respectively. Larger-sized circles reflect larger numbers of publications and thicker curves

Water Practice & Technology Vol 16 No 1
doi: 10.2166/wpt.2020.099

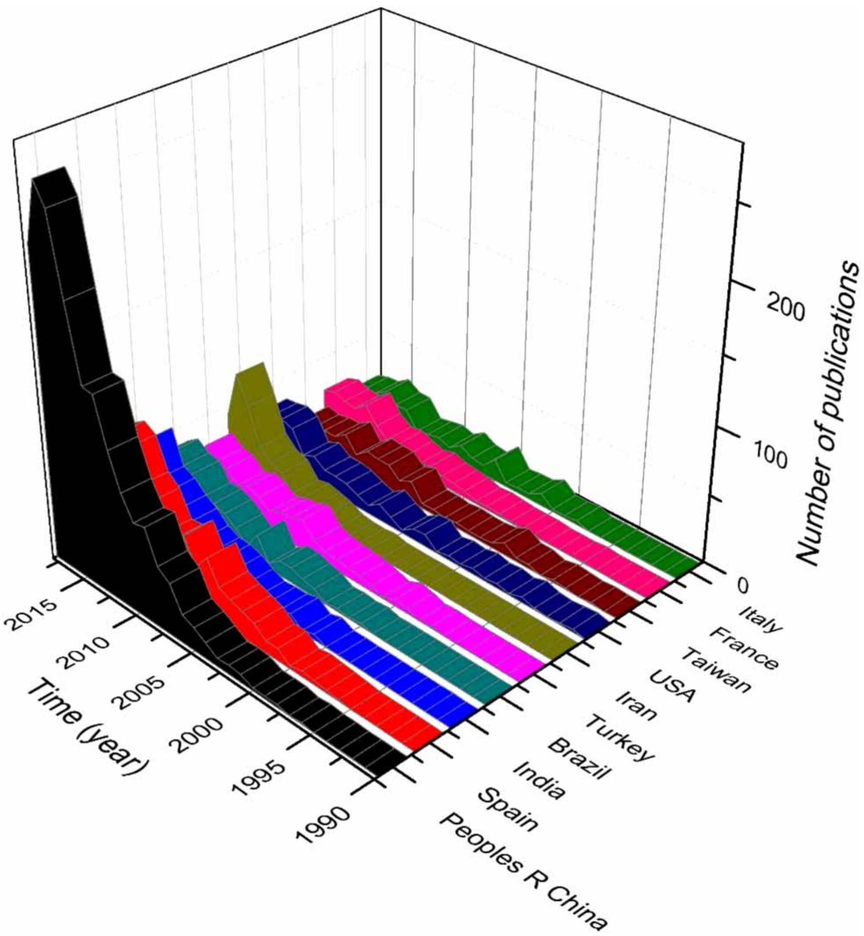

Figure 1 | Growth trends of the number of publications from the top 10 most productive countries.

represent stronger collaborations. The largest circle belonged to the PRC, following Figure 1. The strongest collaboration connection was between the PRC and the USA, but Spain achieved the broadest collaboration with other countries.

Table 3 lists the top 20 most-productive institutions. This table shows that the University of Barcelona was in first place in terms of both numbers of publications and citations. This institution exhibited dominance in the field of the research on Fenton-based AOPs because it is in charge of national research programs, with the UB office of International Research Projects responsible for European projects in this area. It should also be noted that the Chinese institutions occupying second and third places were the Chinese Academy of Sciences and the Harbin Institute of Technology, although their citations were not as numerous as their number of publications. Conversely, while the University of Paris Est was in seventh place, it performed better in terms of citations, with its TLCS and TGCS numbers both in second place.

Journal analysis

To facilitate the researchers working on this subject, journal analysis was conducted. While 5,940 publications were from 746 journals, just 11.26% of journals were responsible for more than ten publications. Figure 3 and Tables 1 contain the data of the 20 most productive journals and their total citations. As shown in Figure 3, the most productive journal was the Journal of Hazardous Materials. Although the number of publications from the second-place journal, the Chemical Engineering Journal, was close to that of the first journal, it had about 2.5 times fewer citations than the first journal.

Water Practice & Technology Vol 16 No 1
doi: 10.2166/wpt.2020.099

Table 2 | Top 20 countries publishing research on Fenton-based AOPs during 1990–2019

Country	N[a] (R[b])	TLCS[c] (R)	TGCS[d] (R)
People's Republic of China	1,698(1)	8,268(2)	25,766(2)
Spain	830(2)	10,916(1)	34,531(1)
India	421(3)	3,603(4)	11,153(3)
Brazil	311(4)	2,183(10)	6,028(10)
Turkey	296(5)	2,503(8)	7,105(7)
Iran	289(6)	1,128(13)	3,098(16)
USA	275(7)	2,743(6)	9,781(5)
Taiwan	242(8)	2,891(5)	6,842(8)
France	224(9)	3,626(3)	9,961(4)
Italy	209(10)	2,313(9)	9,672(6)
Portugal	190(11)	2,701(7)	6,719(9)
Malaysia	152(12)	1,081(14)	3,200(15)
Mexico	144(13)	551(20)	2,955(17)
South Korea	139(14)	982(16)	3,477(14)
Greece	101(15)	1,179(12)	3,775(13)
Japan	97(16)	702(17)	2,729(18)
Canada	97(16)	488(21)	2,542(19)
Australia	96(17)	991(15)	5,239(11)
UK	95(18)	649(19)	2,142(20)
Poland	95(18)	337(22)	1,348(22)
Tunisia	93(19)	690(18)	1,684(21)
Germany	87(20)	1,567(11)	4,564(12)

[a]Total number of publications.
[b]Ranking in corresponding column.
[c]Total local citation score.
[d]Total global citation score.

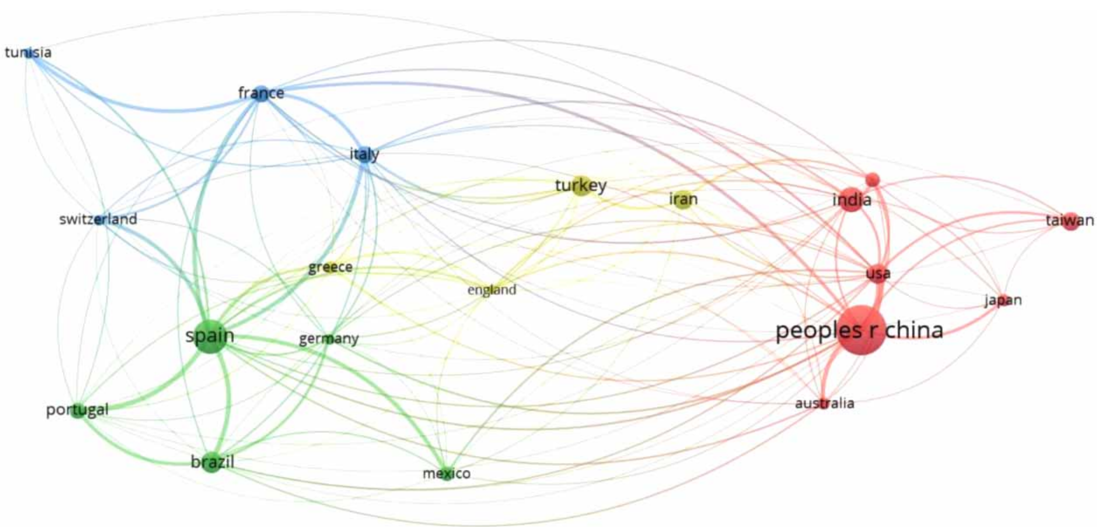

Figure 2 | Collaboration network between the top 20 countries.

Water Practice & Technology Vol 16 No 1
doi: 10.2166/wpt.2020.099

Table 3 | Top 20 most productive institutions during 1990–2019

Institution	Recs	TLCS[a]	TGCS[b]
Univ Barcelona	171(1)	3,819(1)	12,454(1)
Chinese Acad Sci	153(2)	1,216(6)	4,016(6)
Harbin Inst Technol	116(3)	575(11)	1,758(11)
Univ Porto	113(4)	1,758(3)	4,219(4)
Univ Almeria	101(5)	1,562(4)	5,027(3)
Plataforma Solar Almeria	92(6)	1,374(5)	4,132(5)
Univ Paris Est	70(7)	2,249(2)	5,180(2)
CIEMAT	69(8)	947(7)	3,749(7)
Islamic Azad Univ	68(9)	192(22)	602(23)
Univ Sao Paulo	64(10)	377(17)	943(20)
Chia Nan Univ Pharm & Sci	62(11)	598(10)	1,472(12)
Ecole Polytech Fed Lausanne	58(12)	565(12)	1,900(9)
Tsinghua Univ	58(12)	348(18)	1,073(16)
Univ Castilla La Mancha	58(12)	912(8)	2,643(8)
Tongji Univ	57(13)	413(16)	1,226(15)
Univ Tabriz	56(14)	496(14)	1,237(14)
Wuhan Univ	55(15)	798(9)	1,845(10)
Univ Malaya	50(16)	297(19)	895(21)
Univ Chinese Acad Sci	48(17)	101(23)	420(24)
Nankai Univ	46(18)	496(14)	866(22)
Istanbul Tech Univ	45(19)	228(20)	1,095(19)
Univ Zagreb	44(20)	512(13)	1,157(18)
Zhejiang Univ	44(20)	455(15)	1,181(17)

[a]Total local citation score.
[b]Total global citation score.

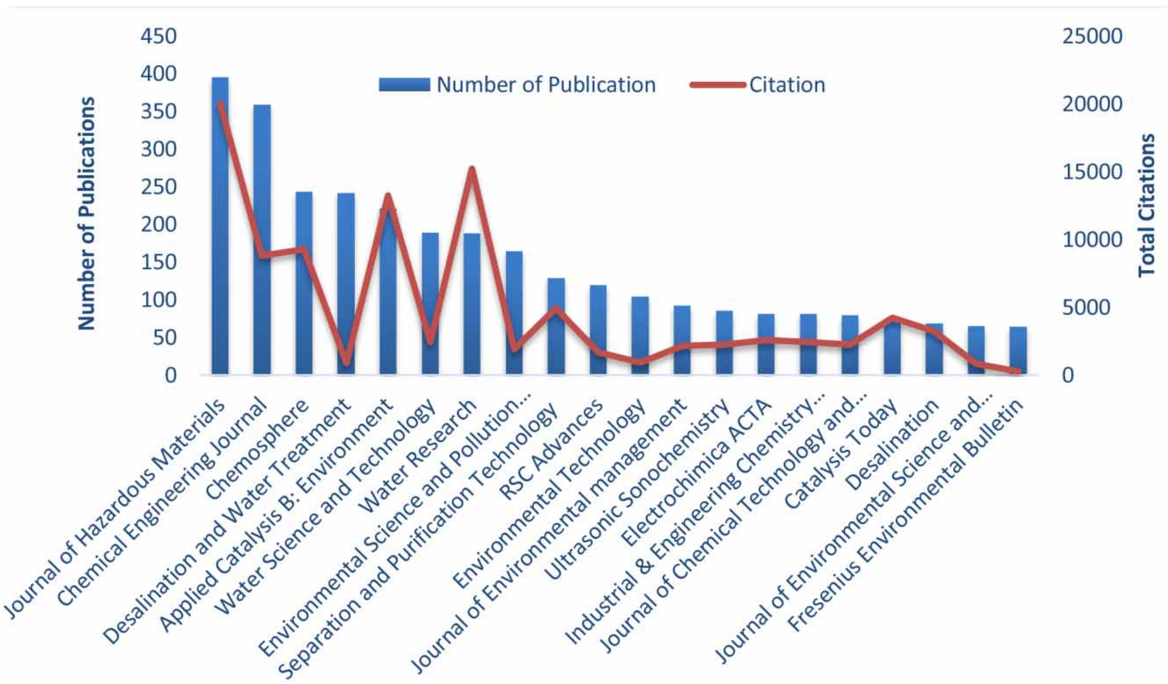

Figure 3 | The 20 most productive journals and their total citations.

Water Practice & Technology Vol 16 No 1
doi: 10.2166/wpt.2020.099

Keyword analysis

13,161 different keywords were identified, with just 886 keywords appearing more than 10 times. Figure 4 shows the co-occurrence network of the most frequently-occurring 100 keywords, with circles and curves are representative of keywords and their co-occurrence relationships. As shown in Figure 4, the keywords were divided into four clusters, identified as follows:

Cluster 1(in red): keywords related to the topic 'textile wastewater'.

Cluster 2(in green): keywords regarding to the topic 'catalyst'.

Cluster 3(in blue): keywords related to the topic 'photo Fenton processes'.

Cluster 4(in yellow): keywords regarding to the topic 'electro Fenton'.

The four clusters revealed the major directions of Fenton-based research. Keyword analysis in Figure 4 showed that textile and pharmaceutical wastewaters are the most studied wastewaters applying Fenton-based AOPs. A more in-depth analysis of these keywords follows.

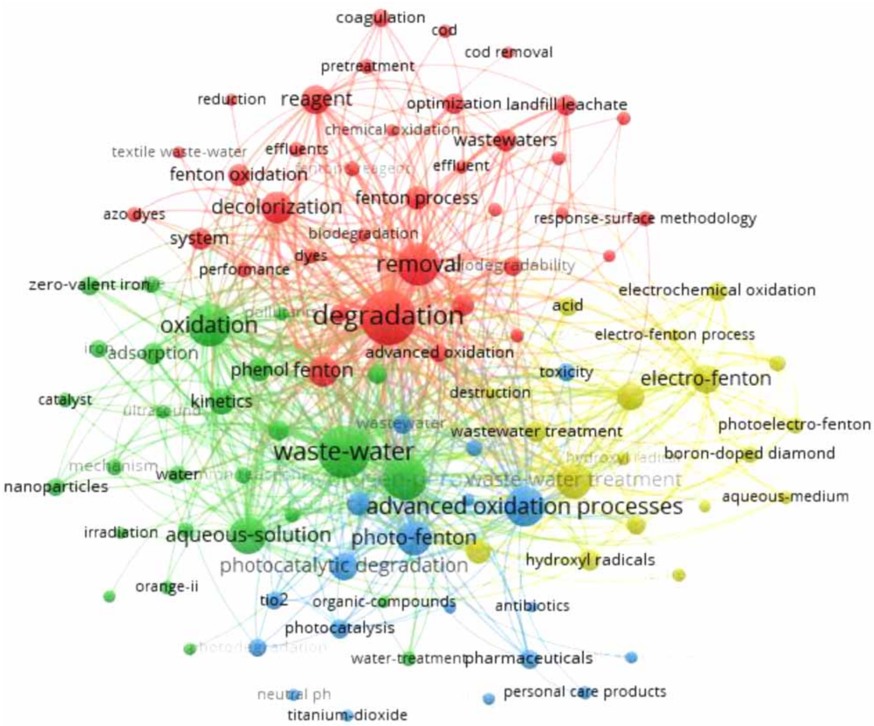

Figure 4 | Co-occurrence network of the top 100 keywords with the most frequent occurrence.

Cluster 1- textile wastewater: The keyword 'textile wastewater' is the core of Cluster 1, revealing one of the leading research directions of Fenton-based processes. This keyword reflected one of the most critical industrial wastewater sources, in which large amounts of textile dyes are discharged into aquatic environments through textile effluents (Asghar *et al.* 2015). Because the discharge of even a small quantity of such dye can produce unacceptable toxic compounds, decolorization from textile wastewater seems to be one of the most challenging tasks faced by environmental engineers (Argun & Karatas 2011). Despite the substantial body of research to improve the level of fixation of dyestuffs onto the substrate, color pollution in aquatic environments is still an escalating problem (Pearce *et al.* 2003). The keyword 'azo dyes' reflects the most important contaminant in textile wastewaters. The adverse effects of azo dyes in the environment, such as their inhibitory effect on aquatic photosynthesis, their capability to deplete dissolved oxygen (DO), and their toxicity to humans and most kinds of ecosystems, are principal concerns related to these contaminants (Argun & Karatas 2011). Because of their complex structure and electron-withdrawing capacity, azo dyes are resistant to

Water Practice & Technology Vol 16 No 1
doi: 10.2166/wpt.2020.099

biodegradation (Dave *et al.* 2014), so decolorization based on Fenton-based processes has attracted many researchers involved in related studies. Some other industrial wastewaters include pharmaceutical residues, phenols, and pesticides containing high COD, which are not biodegradable, so pretreatment or treatment with Fenton-based processes also represents an alternative method for their COD removal (Capodaglio *et al.* 2018; Ganiyu *et al.* 2018).

Another keyword in this cluster is 'landfill leachate'. Generated leachate like textile wastewaters contains refractory contaminants. It has high COD, BOD5, and associated hazard potential. While biological treatment technologies alone are not effective for the degradation of leachate, Fenton based processes have been detected as efficient treatment methods that lead to rapid degradation of refractory contaminants in landfill leachates (Biglarijoo *et al.* 2016).

Cluster 2- catalyst: The keyword 'catalyst' is the core of this cluster. As illustrated in Table 1, Fenton-like or catalytic Fenton, sometimes called heterogeneous Fenton, are related to Fenton-based processes in that they also use solid catalyst sources such as zero valent iron (ZVI), zero valent aluminium, and other particles rather than soluble iron salts for the decomposition of $H_2O_2$to form HO• (Ganiyu *et al.* 2018). Choice of the best catalyst offering high performance at low cost has been a hot topic in this area for decades.

The keyword 'zero valent iron' is related to the most important catalysts used in Fenton-like processes. ZVI is an emergent material with several advantages. It is widely available, cheap, simple, and easy to handle, and the technology is straightforwardly scalable (Raji *et al.* 2020). ZVI technology is based on the use of iron in its elemental state and the material can be used at the microscale (powders) or nanoscale, or in the form of wires, nails, and wool (Zhang *et al.* 2019).

Keywords 'adsorption' and 'kinetics' can be related to catalytic degradation technologies. The results from many studies obviously indicated that a heterogeneous catalyst is more efficient than homogeneous catalysts because of the 'adsorption' of the contaminant molecules on its surface (Litter & Quici 2011). The kinetic rate constant of pollutant degradation has also been investigated in many studies (Lathasree *et al.* 2004). The keyword 'phenol' is also indicative of the more frequently-found target contaminants in Fenton-based processes. It has been reported that catalyst-based AOPs are effective for the removal of various contaminants such as 4-chlorophenol and phenol (Bokare & Choi 2014). Phenolic compounds threaten the environment because of their high toxicity and the fact they can remain in the ground for long periods of time (Morshed *et al.* 2019).

Cluster 3- photo Fenton processes: The keyword 'photo Fenton' is the core of this cluster, and nearly all keywords in this cluster are related to this topic. The keywords 'photo Fenton', 'photocatalytic degradation', 'photodegradation', and 'solar photo-Fenton' appearing in this cluster are common processes that use photo reduction and photolysis for pollutant degradation. While photo Fenton and solar photo-Fenton are two kinds of Fenton-based AOPs, and a photocatalytic process is usually compared with these two processes in studies, it can also be employed together with Fenton-based AOPs. A photo-Fenton process uses a combination of Fenton reagents and UV-Vis radiation ($\lambda < 600$ nm) that energetically improves the Fenton process by giving rise to extra HO• radicals (Rahim Pouran *et al.* 2014), with cost one of its main limitations. Several strategies, primarily the application of heterogeneous (photo) catalysts and/or chelating agents, have been used to minimize cost and improve photo-Fenton efficiency. Some heterogeneous photo-Fenton catalysts used for degradation of recalcitrant organic compounds are $BiFeO_3$, Fe-zeolites, LiFe $(WO_4)_2$, and Fe(III)–SiO_2 (Farzadkia *et al.* 2015). A solar photo-Fenton process uses sunlight rather than artificial light for the photo-Fenton reaction to dramatically reduce process cost (Malato *et al.* 2002). Semiconductor catalysts like TiO_2, Fe_2O_3, ZnO, CdS, and ZnS, applied for heterogeneous photocatalysis, have proven effective in degrading several refractory 'organic compounds' into readily biodegradable compounds, and through mineralization into carbon dioxide and water. Semiconductor photocatalysis is an economical, ecologically-friendly, and sustainable process method used in wastewater treatment (Chong *et al.*

Water Practice & Technology Vol 16 No 1
doi: 10.2166/wpt.2020.099

2010). The keyword 'TiO_2' in this cluster reveals the most attractive semiconductor catalyst that has received the greatest attention in photocatalysis technology because it remains stable after repeated catalytic cycles (Pérez *et al.* 2018).

Another keyword in cluster 3 is 'pharmaceuticals'. Pharmaceutical industries produce wastewater containing toxic solvents and recalcitrant compounds (Rahim Pouran *et al.* 2014), and since many studies have described the limited effectiveness of many conventional treatment technologies for non-biodegradable pharmaceutical wastewaters, AOPs have demonstrated great capability for degradation of many recalcitrant pharmaceuticals (Rahim Pouran *et al.* 2014). Among AOPs, a homogeneous photo Fenton process has been reported as one of the most appropriate methods (Rahim Pouran *et al.* 2014). The keyword 'antibiotics' in this cluster represents the most-studied pharmaceuticals that are most highly consumed and thereby cause major problems in aquatic environments even in small quantities because their resistant behavior may give rise to antibiotic-resistant bacteria (Pirsaheb *et al.* 2018).

Cluster 4- electro Fenton processes: The keyword 'electro Fenton' is the core of cluster 4 and most keywords in this cluster are directly or indirectly relevant to this topic. The keyword 'electrochemical oxidation' in this cluster is related to recent emerging technologies related to the elimination of a variety of contaminants from aquatic environments (Brillas *et al.* 2009). This process can occur in electrolytic cells either by a direct electron transfer to the anode (including conventional procedures of cathodic reduction and anodic oxidation) or by a chemical reaction at the electrode with electro-generated species from pollutants. Most electrochemical technologies are based on indirect electrolysis in which a solution's target contaminant is eliminated by reversibly or irreversibly electro-generated reagents at the electrode (Tarr 2003). Among these technologies, electrochemical advanced oxidation processes (EAOPs) provide several advantages, such as environmental friendliness because the electron is a clean reagent, safety because they operate at room temperature and pressure, high energy efficiency, easy handling, and versatility (Sirés *et al.* 2014). EAOPs are mediated electrochemical treatments based on either destruction of persistent organic pollutants at the anode or using the Fenton's reagent partially or completely generated from electrode reactions (Brillas *et al.* 2009). The electro Fenton (EF) process, a common and widely-studied EAOP based on Fenton reaction chemistry (Oturan 2000; Brillas *et al.* 2009; Moreira *et al.* 2016), can overcome some of the classical Fenton drawbacks (such as problems associated with the storage and transport of H_2O_2, the use of high amounts of iron and corresponding sludge production) and increase the efficiency of pollutant removal (Moreira *et al.* 2016; Kim *et al.* 2018). It is comprised of (1) the *in situ* electrochemical production of H_2O_2 on the cathode and (2) sacrificial production of Fe^{2+} on the anode or external addition of Fe^{2+} (Brillas *et al.* 2009; Moreira *et al.* 2016; He & Zhou 2017). Photo-electro Fenton (PEF), another kind of electrochemical oxidation, is suitable for additional destruction of organic contaminants from waters, combines the electro Fenton process with irradiation provided by artificial light (Brillas & Martínez-Huitle 2015; Moreira *et al.* 2016). The main drawback of this process is high electrical cost due to the use of artificial lamps, and a process called sono-electro-Fenton (SEF) that uses free and renewable natural solar light has emerged ($\lambda > 300$ nm) (Brillas & Martínez-Huitle 2015; Moreira *et al.* 2016). Since heterogeneous $HO\bullet$ is generated at the anode surface, recent advances of these processes depend on the nature of the anode material, (Brillas & Martínez-Huitle 2015), because the use of efficient anode materials can prevent potential electrode deterioration. Also, by choosing high oxygen overvoltage anodes, hydroxyl radicals can be efficiently produced to enhance treatment efficiency (He & Zhou 2017). Some anodes used in electrochemical processes are Pt, PbO_2, boron-doped diamond (BDD), SnO_2, and TiO_2. The keyword of 'boron-doped diamond' in Cluster 4 reveals the most-studied anode related to electrochemical processes. A BDD anode is most suitable for anodic oxidation because it is non-active and has several important characteristics such as high corrosion stability even in a strongly acidic environment, an inert surface with low adsorption properties, and high O_2 evolution overvoltage (Moreira *et al.* 2016). Besides, its

Water Practice & Technology Vol 16 No 1
doi: 10.2166/wpt.2020.099

remarkable capacity for producing hydroxyl radicals leads to a high level of organic contaminant mineralization (GilPavas *et al.* 2018).

Many previous studies have demonstrated that applying BDD in different EAOPs is excellent in removing dyes from synthetic and industrial effluents (Panizza & Cerisola 2008; Migliorini *et al.* 2011; Sales Solano *et al.* 2013) and oxidizing phenol (Lee *et al.* 2017) and carboxylic acids (Brillas *et al.* 2010; Hui *et al.* 2013). Notably, BDD deposited on several support materials like Nb, Ti and Si has been widely studied for use in dye removal (Brillas & Martínez-Huitle 2015).

Detecting reference with strong citation bursts

Discovery of the most active area of research during a specific period of time can be disclosed by a citation burst applied to detect dramatic increases of interest in a particular specialty; that is, it is an indicator for a cited reference by exhibiting a sharp increase in citation number during a certain period (Chen 2006; Garfield 2009). Burst strength and begin-and-end points are metrics for a citation burst. In 101,127 valid references involved in publishing 5,940 records, 249 citation bursts were found, and Table 4 lists the top 20 references associated with the strongest citation burst. As it can be seen in this table, the study of Brillas *et al.* (2009), a review of electrochemical processes, based on Fenton reaction, received the strongest citation burst. This study discussed the advantages of electrochemical processes based on the Fenton reaction. Interestingly as demonstrated in this table, this study has received the citation burst since 2012. This continuing interest shows that investigating the electro-Fenton process, which is suitable for mineralizing a large variety of pollutants, will not stop, and researchers are still seeking ways to improve this process.

Table 2 shows that this study ranked third in citation number, in contrast to the citation-burst point of view. It is worth noting that the first 14 papers with the strongest citation bursts were review papers, and among them, five papers were related to electro-Fenton processes whose citation histories are shown in Figure 5(a), revealing that electro-Fenton processes received great attention because of their many advantages, as mentioned in keyword analysis section (cluster 4). Table 4 shows the study of Pignatello *et al.* (2006a) to be associated with the second strongest citation burst in the field of organic contaminant degradation, which had received the citation burst in 2011. Three more studies were also related to this area whose citation histories are shown in Figure 5(b). These studies revealed that Fenton-based AOPs were widely used for the degradation of organic pollutants, especially phenol compounds.

Figure 5(a) shows that all references have had a large number of citations since 2011, indicating that electro-Fenton research is still a research focus. However, Figure 5(b) includes references that differ with respect to a citation history view, possibly indicating a resurgence in related research (organic contaminant degradation).

CONCLUSIONS

The time-wise analysis of the research on Fenton-based AOPs reflected positive trends over the last 30 years (1990–2019). The exponential growth is contributed by studies from 59 different countries with PRC and Spain emerging as the countries that have contributed most to research on Fenton-based AOPs. As the majority of these researches are concentrated only in the PRC, there is a huge gap between other countries and China (except for Spain who produced the most cited and significant amount of research in this area), in the production of scientific knowledge on Fenton-based AOPs.

Collaboration by the USA, PRC, Spain, and France with other countries was intensive, with the most intensive collaboration between the USA and the PRC. Based on the journal analysis, the most productive journals in this field was the *Journal of Hazardous Materials*, showing the effectiveness of Fenton-based processes for the degradation of hazardous pollutants.

Water Practice & Technology Vol 16 No 1
doi: 10.2166/wpt.2020.099

Table 4 | Top 20 references with strongest citation bursts

	References	Year	Strength	Begin	End	Topic
1	Brillas E, 2009, CHEM REV, V109, P6570, DOI	2009	91.2943	2012	2019	Electro-Fenton process and related electrochemical technologies based on fenton's reaction chemistry
2	Pignatello JJ, 2006, CRIT REV ENV SCI TEC, V36, P1, DOI	2006	78.2479	2011	2014	Advanced oxidation processes for organic contaminant destruction based on the Fenton reaction and related Chemistry
3	Neyens E, 2003, J HAZARD MATER, V98, P33, DOI	2003	77.6658	2005	2011	A review of classic Fenton's peroxidation as an advanced oxidation technique
4	Babuponnusami A, 2014, J ENVIRON CHEM ENG, V2, P557, DOI	2014	66.1876	2015	2019	A review on Fenton and improvements to the Fenton process for wastewater treatment
5	Sires I, 2014, ENVIRON SCI POLLUT R, V21, P8336, DOI	2014	63.4198	2015	2019	Electrochemical advanced oxidation processes: today and tomorrow. A review
6	Gogate PR, 2004, ADV ENVIRON RES, V8, P501, DOI	2004	54.6993	2005	2012	A review of imperative technologies for wastewater treatment I: oxidation technologies at ambient conditions
7	Bokare AD, 2014, J HAZARD MATER, V275, P121, DOI	2014	52.9465	2015	2019	Review of iron-free Fenton-like systems for activating H_2O_2 in advanced oxidation processes
8	Brillas E, 2015, APPL CATAL B-ENVIRON, V166, P603, DOI	2015	48.3976	2015	2019	Decontamination of wastewaters containing synthetic organic dyes by electrochemical methods. An updated review
9	Nidheesh PV, 2012, DESALINATION, V299, P1, DOI	2012	48.1098	2014	2019	Trends in electro-Fenton process for water and wastewater treatment: An overview
10	Pera-Titus M, 2004, APPL CATAL B-ENVIRON, V47, P219, DOI	2004	45.0594	2005	2012	Degradation of chlorophenols by means of advanced oxidation processes: a general review
11	Oturan MA, 2014, CRIT REV ENV SCI TEC, V44, P2577, DOI	2014	44.4892	2015	2019	Advanced oxidation processes in water/ wastewater treatment: principles and applications. A review
12	Gogate PR, 2004, ADV ENVIRON RES, V8, P553, DOI	2004	40.6021	2005	2012	A review of imperative technologies for wastewater treatment II: hybrid methods
13	Panizza M, 2009, CHEM REV, V109, P6541, DOI	2009	39.5690	2012	2019	Direct and mediated anodic oxidation of organic pollutants
14	Martinez-Huitle CA, 2015, CHEM REV, V115, P13362, DOI	2015	38.3127	2016	2019	Single and coupled electrochemical processes and reactors for the abatement of organic water pollutants: a critical review
15	Chamarro E, 2001, WATER RES, V35, P1047, DOI	2001	36.7109	2002	2009	Use of Fenton reagent to improve organic chemical biodegradability
16	Kang YW, 2000, WATER RES, V34, P2786, DOI	2000	36.1786	2002	2008	Effects of reaction conditions on the oxidation efficiency in the Fenton process
17	Malato S, 2009, CATAL TODAY, V147, P1, DOI	2009	34.3725	2011	2018	Decontamination and disinfection of water by solar photocatalysis: Recent overview and trends
18	Perez M, 2002, WATER RES, V36, P2703, DOI	2002	33.9268	2004	2010	Fenton and photo-Fenton oxidation of textile effluents
19	Xu LJ, 2012, ENVIRON SCI TECHNOL, V46, P10145, DOI	2012	34.7568	2016	2019	Magnetic Nanoscaled Fe_3O_4/CeO_2 Composite as an efficient fenton-like heterogeneous catalyst for degradation of 4-chlorophenol
20	Lucas MS, 2006, DYES PIGMENTS, V71, P236, DOI	2006	33.6551	2009	2014	Decolorization of the azo dye Reactive Black 5 by Fenton and photo-Fenton oxidation

Water Practice & Technology Vol 16 No 1
doi: 10.2166/wpt.2020.099

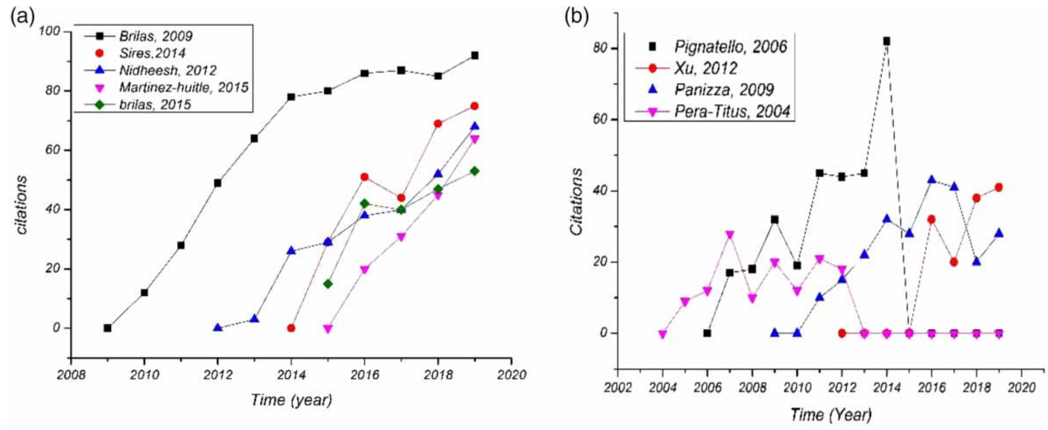

Figure 5 | Historical citation graph for references with strong citation bursts related to the top 2 topics: (a) electro-Fenton, (b) organic contaminant degradation.

Based on the bibliometric analysis, a detailed content analysis had been carried out through most-occurring keywords. It has been observed that among industrial wastewaters, dyes and phenols are the most commonly used pollutants that can be efficiently degraded by Fenton-based AOPs. On the other hand, electro-Fenton, photo-Fenton, and Fenton-like processes were a hot topic on Fenton-based AOPs in the past 30 years. Finally, from the citation-burst point of view, it was found that electrochemical processes, organic pollutants degradation (especially phenolic compounds), photo-Fenton, Fenton-like and textile wastewaters were the hot topics of top references, indicating good agreement with the research topics analyzed through keywords.

These findings showed that hybrid methods involving Fenton-based processes, adsorption, ultrasound, nanofiltration, and many other treatment technologies are increasing and these new approaches can develop effective and sustainable treatment technologies. It is expected that future advances will be performed at the pilot scale, and combined Fenton-based approaches could also be used for wastewater treatment in the industry. Studies about innovative (nano) catalysts, electrocatalytic and photocatalytic materials and also their catalytic mechanisms can provide new degradation possibilities and overcome the Fenton process's drawbacks.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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