



Review

Sonoelectrochemical processes for the degradation of persistent organic pollutants

Wei Lun Ang^a, Patrick J. McHugh^b, Mark D. Symes^b

^a Department of Chemical and Process Engineering, Faculty of Engineering and Built Environment, Universiti Kebangsaan Malaysia, 43600 Bangi, Selangor, Malaysia

^b WestCHEM School of Chemistry, University of Glasgow, Glasgow G12 8QQ, UK

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ABSTRACT

The combination of electrochemistry and ultrasonic irradiation (sonoelectrochemistry) has gained increasing attention in recent years as a method for removing dissolved pollutants from water. This interest stems from the potential for sonoelectrochemical approaches to completely mineralise dissolved pollutants, converting them into harmless mineral species such as water and carbon dioxide. In many cases, the electrochemical and ultrasonic inputs into a sonoelectrochemical pollutant degradation process are found to be synergistic, producing a faster rate of degradation than that produced by the sum of the purely electrochemical or purely sonochemical inputs on their own. This synergism has several causes, with enhanced production of powerfully oxidising radicals and improved mass transport to and from the electrode surface being the two most often cited. In this review, we first give an overview of the various factors that impact on sonoelectrochemical pollutant degradation studies (including reactor design, sonochemical and electrochemical parameters and reaction conditions), before then discussing in detail examples of sonoelectrochemical processes for the degradation of persistent organic pollutants in water from 2015 onwards.

1. Introduction

As industry in the 21st century continues to expand, the need for effective methods for wastewater decontamination has become ever-more pressing. A large number of industrial processes produce and discharge hazardous effluents containing various dyes, pharmaceuticals, pesticides, surfactants, heavy metals, and so on, which pose a potential risk to local ecosystems. Conventional treatment methods have often struggled to deal with this wastewater effectively [1–6]. Persistent organic pollutants, often present in industrial wastewater, are perhaps of the most concern, and have been linked to a number of chronic and acute medical conditions, including cancers, hypertension, cardiovascular disease, diabetes, suppression of the immune system, adverse effects on cognitive and neuro-behavioural function, and disruption of the function of sex steroids and the thyroid gland [7]. It is in this context that Advanced Oxidation Processes have come to the fore for the treatment of such wastewater. Advanced Oxidation Processes generate highly oxidising radical species that are capable of the complete mineralisation of dissolved organic pollutants (i.e. their conversion to ubiquitous and harmless mineral species such as CO₂, water and N₂) [8]. First defined in 1987 as a process which generated hydroxyl radicals in sufficient

quantity to purify water, the definition of an Advanced Oxidation Process has since been expanded to cover the generation of other reactive oxygen species (e.g. sulfate radicals, superoxide radicals etc.) [4].

One effective route to radical generation in liquids is through the application of ultrasonic radiation. When ultrasound is applied to a liquid medium, it gives rise to pressure differentials. Due to the sinusoidal nature of the applied ultrasonic field, points in the medium may be subject to successive periods of positive pressure (compression) and negative pressure (rarefaction) (Fig. 1). During periods of rarefaction, gaseous bubbles form which can increase in size with successive cycles; this process occurs on the microsecond timescale and culminates in violent bubble collapse. These collapses (or “cavitations”) are extremely high energy events and can yield temperatures and pressures calculated to exceed 2000 K and 500 bar [9–11]. The volatilised molecules within these bubbles are subject to such harsh conditions during cavitation that their molecular bonds can be cleaved, yielding radicals [12–16]. In addition, the operating parameters of the ultrasonic irradiation (e.g. frequency, power) can be tuned so as to maximise cavitation, allowing the optimisation of radical production [17]. Furthermore, it has been shown that application of an ultrasonic field to a liquid medium may lead to enhanced mass transport, thermal variations caused by

E-mail addresses: w1_ang@ukm.edu.my (W.L. Ang), mark.symes@glasgow.ac.uk (M.D. Symes).

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cavitation, and shear forces which can affect large molecules, particles, and surfaces [15]. Electro-oxidation of aqueous solutions is also known to bring about generation of hydroxyl radicals ($\cdot\text{OH}$) at the anode. With this in mind, it has long been understood that the coupling of ultrasonic radiation with other methods for radical production (especially electrochemical techniques) can be beneficial, with a synergistic relationship observed in many cases [18–21]. Many of the inhibiting factors associated with electrochemistry, such as electrode passivation and mass transport at the electrode–electrolyte interface, are lessened under the action of an ultrasonic field.

In this review, we present an overview of the main parameters affecting sonoelectrochemical processes and an overview of recent significant works in which a combined ultrasonic-electrochemical (or “sonoelectrochemical”) approach to the degradation of persistent organic pollutants has been taken. In doing so, we aim to build on the last comprehensive review of this subject (in 2015) [22], as well as a number of more focussed reviews that have been within this period [23–25].

2. Important factors in sonoelectrochemical advanced oxidation processes

An understanding of electrode materials, reactor design, and operating conditions is vital for optimising the performance of sonoelectrochemical processes and determining aspects such as pollutant degradation efficiency, process feasibility, energy efficiency, and environmental impacts. A few of the most important parameters which directly or indirectly affect the process performance are described below. The reader is cautioned to keep in mind that some of the information below was obtained from purely sonochemical or purely electrochemical processes, rather than combined sonoelectrochemical processes. Where such information is included, its extension and applicability to sonoelectrochemical processes is explained. In the following, we shall focus largely on oxidative degradation of organic pollutants. Electro-reductive routes to pollutant degradation may also be operating in some cases, and interested readers are referred to a recent review of electro-reductive water treatment [26].

2.1. Electrode materials and electrode design

The electrodes are critical to the selectivity and degradation efficiency of electro-oxidation processes. Suitable electrodes/electrode

materials should possess the following characteristics: high stability, chemical/physical resistance to corrosion/passivating layer formation, high electrical conductivity, good catalytic activity and selectivity, low cost/life ratio, to name a few [23]. The materials used to fabricate the electrodes (especially the anode) should also have a high overpotential for water oxidation, which will help to reduce parasitic oxygen generation and hence improve organic pollutant degradation efficiency. Electrodes can be sorted into two classes: active electrodes and non-active electrodes. Active electrodes possess a strong interaction between the electrode surface and hydroxyl radicals, which leads to partial (selective) oxidation of organic pollutants. Examples of materials used for the fabrication of active electrodes are platinum, ruthenium oxide, iridium oxide, and carbon-based materials [23,27]. On the other hand, the interactions between the electrode surface and hydroxyl radicals are much weaker for non-active electrodes (e.g. boron-doped diamond, lead oxide, tin oxide), favouring complete mineralisation of pollutants and enabling degradation of pollutants in bulk solution too [22,23,27].

There are several challenges associated with both classes of electrodes that obstruct their practical application in real systems. For instance, lead oxide electrodes have a relatively short service life due to surface corrosion, and concerns of the possible release of toxic Pb(IV) ions have further reduced its attractiveness [28]. Boron-doped diamond electrodes are costly and economically infeasible for large-scale production [27]. For active electrodes, cost also inhibits the practical application of platinum electrodes, on top of its limited ability to completely mineralise pollutants as compared to non-active electrodes [29,30]. Carbonaceous anodes such as graphite are vulnerable to surface corrosion and the use of other metal oxide materials as pure electrodes is cost inefficient [23,31–33]. All these challenges drive researchers to look for alternative electrodes with satisfactory degradation performance, yet which are stable and economically feasible. Mixed metal oxide electrodes, also known as dimensionally stable anodes, are one popular class of electrode that has captivated interest for sonoelectrochemical processes [32,34]. Typically, mixed metal oxide electrodes consist of an inert substrate (titanium, carbon, stainless steel, etc.) covered by a thin conductive layer of a metal oxide or mixed metal-oxides of tin, iridium, ruthenium, titanium, antimony, etc. [23,30–32,34,35]. The coating materials either give the electrode a higher overpotential towards the oxygen evolution reaction, or they improve the electrode’s stability/corrosion resistance, or impart better conductivity, or act in more than one of these ways. Due to these improvements, studies related to mixed metal oxide electrodes have increasingly attracted the attention of

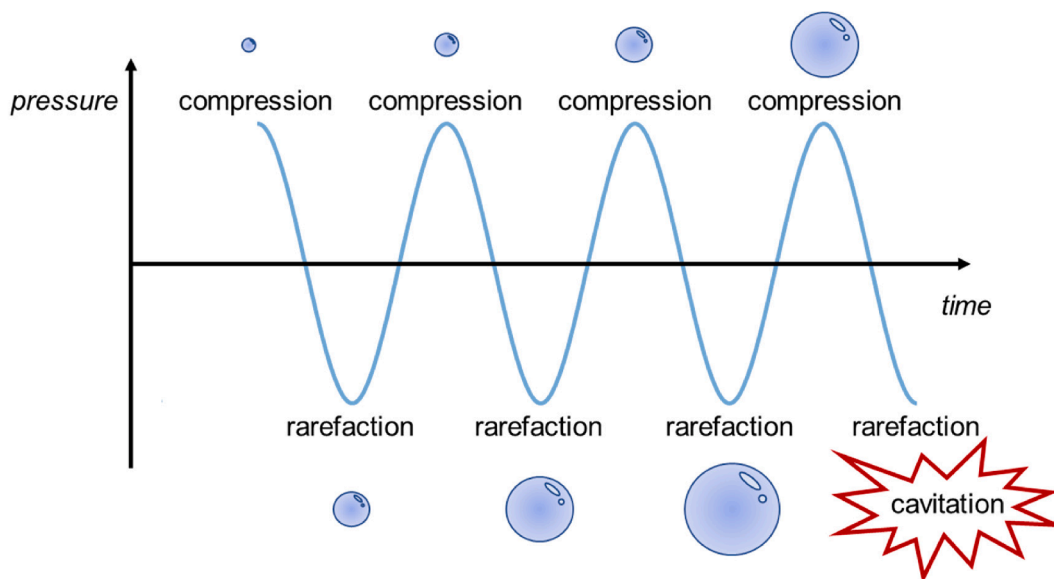


Fig. 1. Diagram of bubble growth and cavitation under the action of a sinusoidal ultrasonic field.

researchers. Mixed metal oxide anodes can be categorised into two classes: bulk mixed metal oxides and supported mixed metal oxides [34]. In bulk mixed metal oxides, the metal oxides are coated on the substrate electrode and provide active sites for electrocatalytic reactions. For instance, the coating of ruthenium-iridium oxides on titanium electrodes has been shown to enhance the sonoelectrochemical degradation of malachite green dyes from 66 to 94% as compared to pristine titanium electrodes [33]. On the other hand, supported mixed metal oxide anodes have multi-layered structures, typically an active oxide layer, a dispersion layer, and a supported oxide layer coated on the substrate electrode. Although the supported oxide layer does not directly contribute to the oxidation of the pollutants or oxygen evolution, such a synthesis strategy can improve the electrode's conductivity and the properties of the surface layer (e.g. electrocatalytic, mechanical, chemical stability, surface area, and service life) for better degradation performance [36–39]. For example, antimony-doped tin oxide was coated as a supported oxide layer on titanium for the electrodeposition of a lead oxide active layer [39]. It provided larger surface area and a greater number of active sites for the loading of the lead oxide, which improved the electrochemical activity of the mixed metal oxide anode for dye degradation.

Innovation in the synthesis and modification of mixed metal oxides has also become a popular area of study. For example, Yang et al. [40] compared the performance of mixed metal oxide anodes prepared via different synthesis approaches when treating cephalosporin pharmaceutical wastewater. A traditional mixed metal oxide anode was prepared by coating ruthenium oxide on titanium sheets while a nano-coated mixed metal oxide electrode was fabricated by brushing ruthenium and iridium nanoparticles (produced via a sol–gel method) onto the titanium plate. Interestingly, both electrodes produced roughly the same amount of hydroxyl radicals when there was no sonication. With ultrasound being applied, the nano-coated electrode system produced 35% more hydroxyl radicals than the system with a traditionally-synthesised dimensionally-stable anode. The authors postulated that the nanostructured mixed metal oxide offered a higher surface area and that the radicals generated by the nano-coated electrode could rapidly diffuse into solution when an ultrasonic field was applied, giving access to the production of more radicals to react with contaminants in the wastewater. Shestakova et al. [41] demonstrated that the electroactivity of the anode could be manipulated by adjusting the formulation and number of coating layers (comprised of tantalum and tin) drop-casted onto a titanium support. By properly adjusting the formulation and number of coating layers, the highest degradation of methylene blue could be obtained.

Another type of modification that can be performed on mixed metal oxide anodes is doping. The incorporation of dopants (e.g. antimony and cerium) in the mixed metal oxide layers will create defects that increase the number of electron transfer channels, and subsequently improve the conductivity and catalytic activity of the electrode [34]. For example, Xu et al. [42] synthesized a Ti/SnO₂-Sb/Ce-PbO₂ anode and successfully used it in the sonoelectrochemical degradation of perfluorooctanoic acid in water. The electrode was synthesised through a series of steps: titanium sheet was dipped in a solution of tin and antimony dopant (prepared via a sol–gel technique), followed by coating of cerium-doped lead oxide on the modified titanium sheet [43]. Meanwhile, Wang et al. [44] showed that doping a titanium electrode coated with tin and ruthenium oxide layers with 1.5% ytterbium increased the oxygen evolution potential and electrocatalytic (organic) oxidation activity of the anode compared to the undoped electrode. All these examples highlight that there is room for improvement in electrode materials, formulations, and synthesis approaches that will enhance their activity for sonoelectrochemical processes. Interested readers are recommended to refer to other reviews which have summarised the literature on electrode materials for purely electrochemical reactions, which might also be of relevance to sonoelectrochemical processes [5,31,32,34,45–51].

In terms of areas for future development in electrode materials for

sonoelectrochemical processes, inspiration from purely electrochemical systems is plentiful. Magnéli phase sub-stoichiometric titanium oxides (with general formula Ti_nO_{2n-1}) are one such emerging electrode material. For example, Ti₄O₇ has been used in fuel cells and batteries due to its wide potential window, high oxygen evolution overpotential, strong corrosion resistance, and great conductivity [32], all useful features for sonoelectrochemical water treatment processes.

Static, two-dimensional parallel-plate electrodes have been frequently used in sonoelectrochemical studies due to the ready availability of such materials and their potential for scale-up. However, other electrode geometries and configurations such as cylindrical and rotating electrodes as well as three-dimensional electrodes should also be explored since these designs could increase the mass-transport coefficient and surface area for radical production and degradation of pollutants as reported in purely electrochemical studies [52]. The influence of ultrasound on the electrodes in these alternative configurations has never been investigated for sonoelectrochemical processes, which thus represents a key area for future study on electrode materials. As one such example, systems employing bipolar bed-type approaches where the gap between the electrodes is filled with conductive particles have the potential to be highly effective for pollutant removal from wastewater (see Fig. 2) [53]. In such a design, the conductive “particle electrodes” are polarised, giving many charged microelectrodes that serve to enhance the mass transfer efficiency of the pollutants, increase the surface area for oxidation, and subsequently endow the entire system with better pollutant removal efficiency. Commonly used particle electrodes include granular activated carbon and carbon aerogels [54–57]. These particles can be modified to enhance their contribution to the degradation efficiency, as reported by Kang et al. [58] where bamboo biochar impregnated with titanium and tin particle electrodes performed better than bamboo biochar alone. The removal rate of the chemical oxygen demand and dissolved organic carbon in coking wastewater using the modified biochar system reached 72% and 65%, respectively, 15–30% better pollutant degradation metrics than for the corresponding electrochemical system using pristine biochar. Other non-conventional particle electrodes such as aluminium oxide embedded with titanium-tin-antimony and conductive polyurethane/polypyrrole/graphene have also been developed to aid the electrochemical degradation of pollutants [59,60]. This reflects the flexibility and versatility of material development for such electrodes. However, particle electrodes have not yet been applied for sonoelectrochemical processes, suggesting a promising avenue for future development.

2.2. Reactor design and modes of operation

Most of the published articles on sonoelectrochemical processes employ ultrasonic bath type emitters while the majority of the remainder utilise ultrasonic immersion horn probes. Experimental configurations as shown in Fig. 3 describe the typical set-ups used in sonoelectrochemical processes for pollutant degradation. For ultrasonic bath type emitters, a transducer that converts electricity into an ultrasonic vibration to generate the sound waves is attached to the bottom of the ultrasonic bath. Normally, the working solution is separated from the ultrasonic bath and the sound waves must penetrate through the bath solution and electrochemical reactor wall before reaching the working solution and electrodes. The intensity of bubble production and the formation of reactive radicals are expected to be reduced as a result. Nonetheless, this configuration enables the acoustic energy field to be reasonably evenly distributed throughout the reactor, giving an equal impact on and across both electrodes [22].

For ultrasonic horn type emitter configurations, the ultrasonic horn is immersed in the working solution together with the electrodes. The cavitation intensity is at its highest around the horn, and it fades in strength when moving further away from the horn tip. Hence, the electrodes must be placed close to the ultrasonic horn to enjoy the benefits of intense mixing, electrode cleaning, and generation of reactive

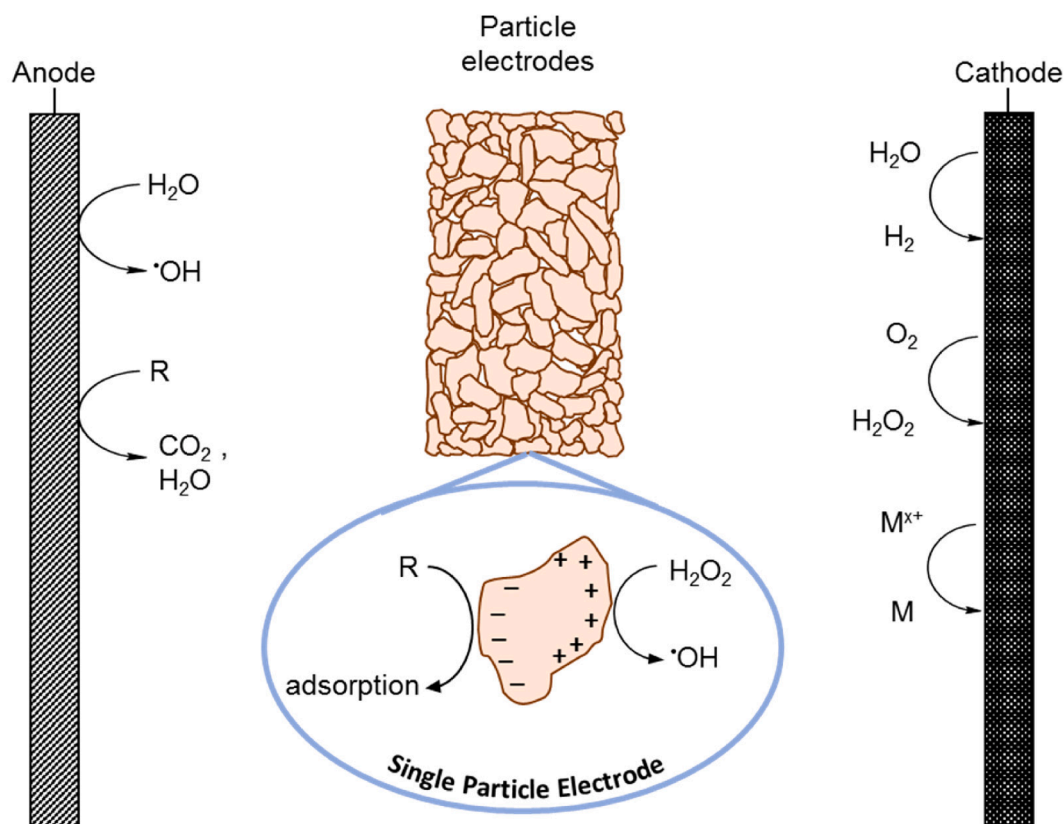


Fig. 2. A reactor-bed type electrochemical reactor design, with an inset showing how each particle can act as a bipolar electrode when under the influence of the electric field applied between the anode and cathode (adapted from ref. [53]).

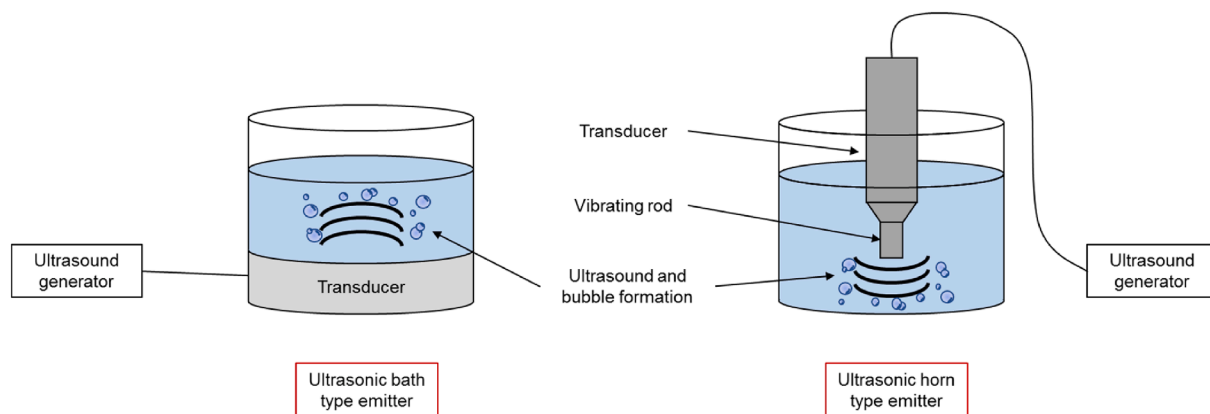


Fig. 3. Bath vs. horn ultrasonic reactor configurations, adapted from ref. [22].

radicals [22,23]. However, due to the intense ultrasonic fields at the tip, tip erosion can contaminate the system and reduce the efficiency of the horn. A much less explored ultrasonic emitter design is the *sonotrode*, where the titanium ultrasonic horn probe itself acts as both an ultrasound emitter and the working electrode. Although the concept might provide a less bulky design compared to ultrasonic bath or horn emitters and separate electrodes, the need for exact potentiostatic control has made it difficult to apply sonotrodes in most sonoelectrochemical processes to date (for example, maintaining a robust electrical contact during operation can be an issue). This explains why only a handful of studies have been reported using a sonotrode design [61–63].

Focussing just on the use of ultrasonic baths and horns, comparative studies as to which approach is more effective are few. A small number of studies have employed both ultrasonic bath and horn configurations

in their experimental set-ups, but the intention in these cases was less to compare the pollutant degradation performance engendered by the use of baths or horns [64–68], but instead to use the horn emitter for low frequency operating conditions while the ultrasonic bath was employed for higher frequency operation. The only close comparison found for pollutant degradation was reported by Gogate et al. [69] (albeit the system in question was purely sonochemical) where the authors presented the performance of formic acid degradation in terms of cavitation yield. Cavitation yield is defined as the extent of degradation in moles divided by the volume of the working solution and the power density. The introduction of this term allows a fair comparison between sonochemical processes operating in different working solution volumes, power densities, and ultrasound fields. Gogate et al. [69] observed that the cavitation yield of the sonochemical reactor equipped with an

ultrasonic bath was around 90% higher than the system with an ultrasonic horn, which indicated that the former system has a better process efficiency due to the more uniform energy dissipation over a wider area.

Literature has clearly demonstrated that the use of ultrasound could enhance the degradation of organic pollutants in electrochemical processes. On one hand, ultrasound brings multiple benefits to the system, such as cavitation bubbles that produce more hydroxyl radicals and help to resolve electrode passivation issue [22–24]. On the other hand, the acoustic streaming and micro jetting produced by ultrasound could disrupt the directional movement of charged ions to the electrodes and disrupt reaction at the electrode. Also, an excessive number of bubbles could reduce the conductivity of the working solution, reducing the synergistic effect of the sonoelectrochemical process for pollutant degradation [70,71]. All these could contribute to unsatisfactory performance and low energy efficiency. Considering these concerns, the influence of ultrasound operated in pulse mode has been investigated [72]. Donoso et al. [73] provided insight into the influence of the pulse-pause ratio on degradation performance during a purely sonochemical process. The use of pulsed ultrasound gave rise to transient cavitation that was highly efficient for generating oxidising radicals [74]. The transient cavitation was influenced by the duration of pulses and pauses: if the pulses of sonication were short in comparison with the pause periods, the implosion of transient bubbles occurred to a limited extent due to insufficient build-up of pressure, leading to less radical generation and lower degradation performance. In this case, a pulse-pause ratio of 1:1 (i.e. a 100 ms pulse followed by 100 ms of quiet time) was found to give only 18.4% removal efficiency of tartrazine compared to 28.2% when the sonication was operated with a pulse-pause ratio of 5:1 (250:50 ms) [73].

The influence of pulse mode sonication on energy consumption and pollutant degradation by sonoelectrochemical processes has also been explored [75,76]. A dual pulse sonoelectrochemical process was conducted with alternating pulses of ultrasound and electricity (for the electrochemical input). Interestingly, the dual pulse operation mode gave slightly better degradation of *p*-nitrophenol (94%) compared to the sonoelectrochemical process operated continuously (89%) [76]. The authors attributed this improvement to the diminished undesired interference of the ultrasound on the electrochemical process (as described above). More appealingly, the energy use of the dual pulse sonoelectrochemical process was 50% lower than the process running continuously over the same time duration. Another potential energy saving is due to the fact that the pulse method does not give such excessive heating of the solution, minimising the energy consumption for cooling in practical applications. Despite these promising results, pulse mode operation has received much less attention compared to other operating parameters. Further investigation should be conducted since this could help to optimise the energy consumption of

sonoelectrochemical processes.

Along with the variation in ultrasound emitters and mode of operation, there is considerable potential for novel reactor design for sonoelectrochemical pollutant degradation, possibly taking inspiration from purely electrochemical designs. For instance, Fan et al. [77] designed a novel rotating multi-electrode reactor for electrochemical oxidation of phenol pollutants. The degradation rate of phenol was 3.2 mg/min at 300 rpm, which was 1.45 times higher compared to the static system. The rotational movement of the electrodes reduced the thickness of the diffusion layer on the anode, which enhanced the mass transfer of pollutants toward the anode surface for degradation. Although the process included no sonochemical element, the fact that the mass transfer coefficient was improved by 110% with a rotating anode compared to a static anode implies that a similar concept could be adopted for sonoelectrochemical processes provided that the technical challenges in incorporating ultrasound are resolved.

Filter-press cells (Fig. 4a) are popularly used for continuous electrochemical reactors where the electrodes are stacked together, and the working solution is then pumped through the cells (e.g. for electro-oxidation of dissolved pollutants) [80,81]. The use of a continuous reactor allows a more accurate study of scale-up effects on electrochemical processes in real-world applications. However, such designs have not yet been adopted for sonoelectrochemical processes, largely due to the difficulty in incorporating ultrasound into such a compact cell reactor. Another interesting reactor design involves the use of a tubular electrochemical continuous reactor to reduce Cr(VI) in wastewater [79]. In the example shown in Fig. 4b, the tubular reactor consisted of a central polished carbon steel rod serving as the cathode, which was surrounded by a spiral helicoid wire of the same material (as the anode). 40 kHz transducers were attached to the top of the reactor to generate ultrasound for the system. This sonoelectrochemical reactor could be further explored for the degradation of organic pollutants by changing the electrode materials.

The attachment of the transducer is not limited to the bottom or top of the reactor. Rodriguez-Freire et al. [82] have shown that transducers of different frequencies can be mounted on the reactor side walls to produce ultrasound for sonochemical processes. In another case, a triple frequency sonoelectrochemical flow cell was developed where transducers of varying frequencies were mounted on the hexagonal cell wall enclosing a central quartz tube with the working solution flowing inside [69]. The sonoelectrochemical flow reactor showed promising results in degrading formic acid, with good energy efficiency and cavitation yield. Hence it seems that the design of sonoelectrochemical reactors is an area where considerable advances in understanding for the scale-up of sonoelectrochemical process could be achieved, and is therefore an area which should receive more attention.

Ultrasonic wave propagation and cavitation field distribution are

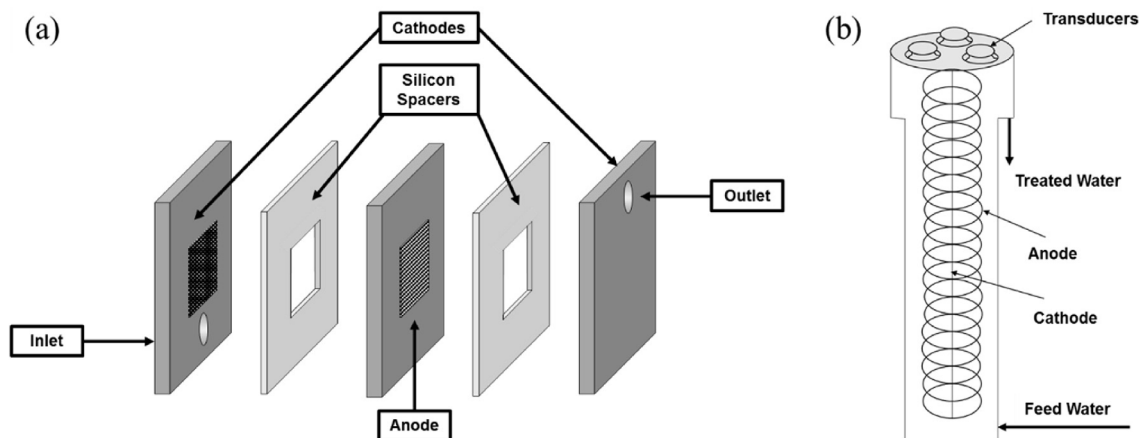


Fig. 4. Reactor designs: (a) an example filter-press cell; (b) a tubular reactor, adapted from references [78] and [79] respectively.

substantially affected by the positioning of electrodes and ultrasound sources. Dietrich et al. [63] highlighted that the insertion of electrodes into a reactor could interrupt ultrasound propagation and affect the sound field and cavitation field distribution, as when the distance of electrodes (without applied potential) was increased from 2 to 3 cm, the degradation of bisphenol A by sonochemical processes decreased. It was postulated that the decrease in distance between the electrodes could increase the interaction of the ultrasound with the electrode surface (reflection/dispersion/absorption), leading to losses and/or interference (enhancement/cancellation) of the acoustic wave [83,84]. On the other hand, when the electrodes were supplied with electricity, shorter inter-electrode distance was found to give higher degradation efficiency due to lower ohmic resistance [63,85]. This agrees with other studies; for example, the degradation of diclofenac increased from 75 to 93% when the distance between the electrodes decreased from 3 to 1 cm [86]. Apart from the resistance factor, another plausible reason for the observed dependence on inter-electrode distance is the distribution of the ultrasound field. For example, Finkbeiner et al. [86] used the chemiluminescence of luminol to show that sonoelectrochemically-generated oxidative species are distributed across the whole volume of the reactor when the inter-electrode distances were set at 1 and 2 cm. However, the oxidative radicals were mostly restricted to the space between the electrodes when the gap between the electrodes was increased to 3 cm, with this reduced cavitation zone contributing to a decline in the degradation efficiency. This clearly indicates the influence of electrode distance on the transmission of the acoustic waves and the shape and size of the cavitation field. Further studies seem warranted here, as a proper understanding of the acoustic field in the reactor as a factor of electrode distance and position will be required for optimisation and scale-up of such sonoelectrochemical degradation processes.

To summarise this sub-section, the development of suitable reactors is essential for optimising and up-scaling sonoelectrochemical processes. As discussed above, ultrasonic propagation and cavitation field distribution are substantially influenced by the geometry of the reactor and the placement of the electrodes. Such factors are further influenced by the height of the working solution, which affects the intensity of the cavitation active zone [87]. Furthermore, the incorporation of ultrasound generators and electrodes into a compact reactor remains a challenge. Therefore, future work should focus on designing sonoelectrochemical reactors that take these factors into consideration.

2.3. Ultrasound power

Ultrasound intensity or power density has an effect on various important properties of sonoelectrochemical processes such as cavitation bubble size, bubble collapse time, cavitation intensity (the implosive energy of the bubbles), and number of cavitation bubbles [22,88]. Typically, an increase in ultrasound power will lead to an improved degradation efficiency of any pollutants present [21,63,89–93]. For instance, Ren et al. [33] reported that the degradation of malachite green improved from 88 to 95% when the ultrasonic power was increased from 100 to 300 W. Similar positive effects have also been observed where the chemical oxygen demand removal efficiency for treating cosmetic wastewater increased from 53 to 62% as the ultrasonic power value increased from 40 to 100 W in a sonoelectrochemical treatment process [94]. All these improvements could be ascribed to the cavitation activity. Under a higher ultrasound power, more cavitation bubbles will be produced, and the collapse intensity will also be enhanced. These phenomena lead to the production of more hydroxyl radicals that in turn promote faster and more complete degradation of pollutants in solution [93]. Furthermore, cavitation creates micro jets and turbulence that provide intense mixing and reduce the extent of any electrode passivation, which together improve the mass transfer of pollutants to the active electrode surface for degradation [40,88,91,93,94].

Nonetheless, operating the sonoelectrochemical system at an ultra-

sonic power exceeding an upper limit will lead to a decline in the pollutant removal efficiency. Several reports have shown that there is an optimal ultrasonic power value (which is case-specific) where the degradation performance peaks before then declining [21,76,95,96]. A few explanations have been given to describe this observation. The repeated and extended effects of cavitation on the electrode surface may in some cases have damaged the electrode and caused the active coating on the electrode to fall off, thus reducing the effectiveness of the electrochemical process [33,97]. On the other hand, the formation of a large number of bubbles in the solution would reduce the transfer of ultrasonic energy to the system due to the scattering of the incident sound by the bubble cloud to the walls of the vessel or back to the transducer [76,94,98,99]. This could be one of the main reasons why the degradation efficiency of pollutants does not continuously improve with increasing ultrasonic power. It is also possible that radical scavenging due to the recombination of hydroxyl radicals reduces the availability of the radicals in the bulk to degrade the pollutants [94,95]. Another plausible explanation is given by Donoso et al. [73] who claimed that at high values of ultrasound power, the bubbles can coalesce to give larger bubbles that then undergo less intense cavitational collapse. Indeed, ultrasound intensity, acoustic power, maximum bubble size and bubble collapse time are all interrelated, as shown in Eqs. (1)–(3): [22]

$$I = \frac{P_A^2}{2\rho c} \quad (1)$$

$$R_m = \frac{4}{3\omega_a} (P_A - P_h) \left(\frac{2}{\rho P_A} \right)^{1/2} \left[1 + \frac{2}{3P_h} (P_A - P_h) \right]^{1/3} \quad (2)$$

$$\tau = 0.915 R_m (\rho/P_m)^{1/2} (1 + P_{vg}/P_m) \quad (3)$$

Where the terms are bubble collapse time (τ), maximum bubble size (R_m), applied acoustic frequency (ω_a), liquid density (ρ), hydrostatic pressure (P_h), acoustic amplitude (P_A), power intensity (I), velocity of sound in the medium (c), vapour pressure in the bubble (P_{vg}), and pressure of the liquid (P_m).

When the ultrasound intensity is high enough, the rarefaction cycle is too short to allow a cavitation bubble to grow to a size sufficient to cause optimal disruption [40,90,97]. Hence, increasing the power substantially above the cavitation threshold would not be beneficial to the sonoelectrochemical process but rather would serve to increase the energy consumption without significantly improving the degradation efficiency.

2.4. Ultrasound frequency

Ultrasound frequency possesses similar influence on sonoelectrochemical processes (such as the number of cavitation bubbles and the violence of cavitational collapse) as ultrasound power, with numerous studies reporting the existence of optimal frequencies for degrading pollutants in water. For instance, the removal efficiency of tartrazine by a sonochemical process was found to peak at 860 kHz at 19.4% as compared to 18.4% and 18% at 578 kHz and 1140 kHz, respectively [73]. Meanwhile, Yang et al. [40] observed that the chemical oxygen demand reduction rate of cephalosporin pharmaceutical wastewater by a sonoelectrochemical process increased from 76% at 25 kHz to around 90% at 45 kHz before declining to 77% when the ultrasound frequency was increased to 85 kHz. However, when a sonoelectrochemical system is operated at sufficiently high ultrasonic frequency (exceeding the optimal level), no optimum point will be obtained. Rather, the lowest frequency adopted in the experimental study will record the best removal efficiency as shown by the study of Shestakova et al. [67] where the highest degradation efficiency (99%) and kinetic rate constant (0.0374/min) of formic acid were obtained after 2 h of sonoelectrochemical treatment at 381 kHz, while increasing the frequency to 992 kHz resulted in a drop in the degradation efficiency and kinetic rate

constant to 91% and 0.0184/min, respectively. All these observations can be explained by the correlation between ultrasound frequency and cavitation activity as summarised by Eqs. (1)–(3).

At a minimum ultrasound frequency of 20 kHz, the number of cavitation bubbles produced is lower than the number of bubbles formed at higher frequencies [100]. With an increase in ultrasound frequency, the number of cavitation bubbles and the number of bubble collapses per unit time are increased and this leads to improved mass-transfer effects (mixing) and the production of more hydroxyl radicals that facilitate the degradation of pollutants. Moreover, it was postulated that the shorter lifetime of bubbles at higher frequencies prevents the recombination of hydroxyl radicals (which produces H_2O_2) through the diffusion of radicals out of the bubbles [19,101]. However, when the ultrasound frequency exceeds a certain threshold level, the degradation rate of pollutants will decline. This phenomenon is similar to the case where the system is operated at very high ultrasound power, where the production of a high number of colliding bubbles with short bubble collapse times results in weakened cavitation intensity, leading to a decline in the production of hydroxyl radicals and the efficiency of pollutant removal [22,40,97,100]. In fact, this is clearly depicted in Eqs. (2) and (3), which show that bubble collapse time is proportional to the maximum bubble size, which is in turn inversely proportional to the applied acoustic frequency. At frequencies above the threshold, the duration of the rarefaction cycle is too short to allow the cavitation bubble to grow to a size capable of releasing sufficient energy upon collapse to generate radicals. Hence, it is vital to operate sonoelectrochemical processes within an optimal range of ultrasound frequency to balance these factors and optimise removal efficiency.

2.5. Current density

Current density is another indisputable parameter that affects the degradation efficiency of pollutants by sonoelectrochemical processes. It is a measure of the amount of electric charge flowing through a unit cross-sectional area of electrode per unit time. This parameter can then be translated into current efficiency and energy consumption that provide an indication of optimal current density. Current efficiency determines the percentage of the charge used in the desired reaction, which in this case is the degradation of pollutants [22]. Two different regimes of operation can be observed depending on the applied current density. When a system is operated below the limiting current density (calculated as shown in Eq. (4)), the kinetics of the reaction are governed by the current, and current efficiencies approaching 100% can be achieved [73]. On the other hand, the degradation process is conditioned by mass transport phenomena when the current density is higher than the limiting current density. In this scenario, the current efficiency will drop below 100% as secondary reactions such as oxygen evolution and electrolyte decomposition take place at the same time [102,103].

$$j_{lim} = 4Fk_m(COD) \quad (4)$$

Where j_{lim} is the limiting current density, F is the Faraday constant, k_m is the mass transport coefficient under hydrodynamic conditions, and COD stands for “chemical oxygen demand”. Chemical oxygen demand can be further defined as the amount of dissolved oxygen that must be present in a given sample in order to oxidise the organic material present. It should be noted that Eq. (4) holds for uniformly accessible electrodes, but non-uniformly accessible electrodes (e.g. porous materials) can give rise to more complex behaviour. The examples that follow show the importance of the current density of sonoelectrochemical processes for optimising degradation performance and current efficiency.

Provided the current density remains below the limiting current density, increasing the current density should enhance the degradation efficiency of pollutants, as shown in various studies such as the decolourisation of amaranth dye (where the removal efficiency was improved from 70 to 90% as the current density increased from 10 to 50 mA/cm^2),

or the decolourisation rate of the dye Cl Reactive Black 8 (which increased from 0.0072 to 0.0133 min^{-1} when the current density rose from 15.9 to 47.6 mA/cm^2) [72,92,104]. This can be explained by the fact that more hydroxyl radicals (which are responsible for pollutant degradation) will be produced per unit time as the current density increases, as long as the current density remains below the limiting value [22,68]. Applying current densities above the limiting current density level will not lead to any further increase in the production of hydroxyl radicals. Instead, the additional charge is passed in performing parasitic water oxidation (to give oxygen), which compromises the current efficiency as part of the energy has been used to oxidise water rather than to oxidise and degrade the pollutants [40,97,105]. This phenomenon is clearly seen in previous studies, where the competing activities at the anode (production of hydroxyl radicals vs. oxygen evolution) resulted in a limiting current density being reached, above which there was no significant improvement in the pollutant removal efficiency [40,94,97]. In other cases, the degradation performance even declined when the employed current density exceeded the optimal value. For instance, the degradation rate constant for sulfamethoxazole pollutants increased as the current density rose from 2 to 20 mA/cm^2 , achieving a maximal value of $2.8 \pm 0.21 \times 10^{-2} \text{ min}^{-1}$, but then declined sharply to only $1.4 \pm 0.14 \times 10^{-2} \text{ min}^{-1}$ when the current density was further increased to 30 mA/cm^2 [106]. These oversupply currents were again seen to cause oxygen evolution at the anode, which disrupted the pollutant degradation pathways. Similar observations have also been reported for the degradation of ofloxacin and perfluorooctanoic acid where sonoelectrochemical processes operated at current densities higher than the optimal values evinced a decline in pollutant degradation rates and efficiencies due to the overwhelming effect of the parasitic oxygen evolution reaction [42,93]. In their system, Xu et al. [42] reported that the energy consumption increased from 55.3 to 89.6 Wh/L when the current density rose from 15 to 20 mA/cm^2 , while the degradation rate simultaneously decreased from 3.1×10^{-2} to $2.9 \times 10^{-2} \text{ min}^{-1}$. This signifies the need to operate the system at an appropriate current density in order to optimise the degradation efficiency whilst also minimising energy consumption and the extent of wasteful side reactions.

2.6. Applied voltage

High cell voltages (generally in excess of 2 V) have to be applied to the electrochemical system for the production of hydroxyl radicals (which are usually the main agents responsible for degrading dissolved pollutants in aqueous solution). Operating the sonoelectrochemical process at high potentials like this leads to high current densities (and hence a large number of free radicals generated per unit time), leading to a higher degradation rate of any dissolved pollutants [107]. Applying high voltages can also help to mitigate against the formation of passivating films on the anode, which can form as the organic pollutants are degraded [63,68,86,90,108–110].

However, the degradation performance of sonoelectrochemical processes starts to decline once the operating voltage exceeds a certain value (which can be case-specific). Just as with current density (to which applied voltage is obviously intimately linked), the decline in degradation rate can be attributed to the occurrence of the oxygen evolution reaction at the anode [75,95,96,111]. For instance, Xie et al. [76] observed that only negligible improvement (<5%) in *p*-nitrophenol degradation was achieved when their cell voltage was increased from 10 to 12 V due to parasitic oxygen evolution. Parsa et al. [112] reported that an increase of applied voltage from 15 to 20 V only improved the removal efficiency of dye pollutants by 7%, but the energy consumption increased from 370 to 570 kWh per kg of chemical oxygen demand removed, clearly demonstrating the importance of the selection of applied voltage for optimal energy consumption.

Considering that the applied voltage has a direct impact on the degradation efficiency of pollutants and the operational cost (energy consumption and lifetime of the electrodes) of sonoelectrochemical

processes, the determination of optimal cell voltage is important. The correlation between the energy efficiency and removal performance can be summarised by Eqs. (5) and (6) below: [22,113]

$$W_{sp} = C_{sp} \times U \quad (5)$$

$$EE = \frac{(c_0 - c_t) \cdot V}{E} \quad (6)$$

Where W_{sp} is the specific energy consumption, C_{sp} represents the specific charge consumption, U is the cell voltage, EE is the energy efficiency, c_0 and c_t are the initial concentration and concentration at a given reaction time, respectively, V is the volume of the solution, and E is the energy consumption.

2.7. Solution pH

Even though pH has a significant effect on the performance of sonoelectrochemical processes, the influence of pH is not always as straightforward to predict as for some other parameters. The pH value affects the mass transfer and production of radicals through changes in surface tension and nucleation that alter the minimum threshold required for the onset of cavitation (the “cavitation threshold”) [114]. The pH can also modify the properties of pollutants by influencing the charge and nature of the interaction between the radicals and pollutants, which can affect the whole degradation process [94,115]. Given that the nature of pollutants varies vastly in terms of charge and hydrophilicity with pH, no general conclusions can be drawn on whether increasing or decreasing pH favours the degradation of pollutants in sonoelectrochemical processes [116]. Nonetheless, a few noticeable theories have been formed in previous studies. One such theory holds that under slightly alkaline conditions, hydroxyl anions in the electrolyte can be oxidized to hydroxyl radicals at the anode surface [96], with this process being enhanced by the presence of ultrasonic waves that drive the anions towards the electrode surface for oxidation [107,117]. In support of this theory, Siddique et al. [117] observed that the degradation of Reactive Blue 19 dye improved by > 40% with an increase of pH from 3 to 8. The subsequent drop in degradation efficiency (by almost 10%) observed when operating this system at pH 9 was explained on the basis that more free radical scavengers such as CO_3^{2-} and SO_4^{2-} could be formed under these conditions, reducing the amount of hydroxyl radicals available for pollutant degradation.

On the other hand, other studies have reported that their sonoelectrochemical processes performed better in a more acidic solution. For instance, the extent of degradation of *p*-nitrophenol and nitrobenzene at pH 3 was much higher than at greater pH values by 30–60% and 2–15%, respectively [75,76]. This was explained on the basis that these acidic conditions disfavoured the competing oxygen evolution reaction [72,75,76], although the performance dropped when the pH was further decreased to pH 2. Under extremely acidic conditions, the authors postulated that the high concentration of hydrogen ions quenched the hydroxyl radicals, thereby reducing the extent of degradation [88,115].

Apart from radical scavenging and the effect on the number of radicals initially produced, the pH might also affect the type of radicals and/or oxidising agents that are produced [80]. For a system utilising chlorine-based electrolytes, hypochlorous acid and hypochlorite will be produced at pH 6–9, generally giving good degradation performance since they are strong oxidants. However, other chloro species such as chlorate and chlorine with poorer oxidation properties will be formed under more alkaline and more acidic conditions [116,118]. This highlights the complex interplay between pH and other parameters that affect the process performance. Hence, the degradation of pollutants is often most optimal within a certain pH range, balancing the benefits of inhibited oxygen evolution and the production of powerfully-oxidising hydroxyl radicals. Stepping outside of this pH range (extremely acidic and/or alkaline conditions), may lead to process performance being

compromised due to radical scavenging and/or competing side reactions.

The second main class of observation concerning pH relates the degradation performance to the properties of the pollutants themselves. In the study conducted by Patidar et al. [93], the removal efficiency of ofloxacin by a sonoelectrochemical process decreased slightly with an increase in pH from 2.7 to 9.9. Apart from the reasons stated above, the authors claimed that such an observation could also be explained by changes in properties of ofloxacin with pH. Ofloxacin is predominantly positively charged (and more hydrophobic) at pH < 6, neutral at 6 < pH < 8, and negatively charged at pH > 8. Hence under acidic conditions, the authors proposed that ofloxacin might accumulate at the interface of the negatively-charged cavitation bubbles (on account of its hydrophobicity) and possibly even diffuse into the cavitation bubbles. Both direct pyrolysis inside the cavitation bubbles and the higher concentration of hydroxyl radicals at the liquid-bubble interface were then held to contribute to a higher degradation efficiency of ofloxacin at pH < 6. This finding aligns with other studies where hydrophobic and/or positively-charged pollutants (e.g. ibuprofen, tetracycline, isoxazolyl penicillins and sulfadiazine) were better degraded under acidic conditions [98,107,114,119–122].

2.8. Electrolyte and electrolyte concentration

The type of electrolytes (salts) in the solution plays an important role in affecting the degradation of pollutants in sonoelectrochemical processes. Some salts such as NaCl, Na_2SO_4 and Na_2CO_3 are capable of being converted into oxidizing agents that lead to indirect oxidation of pollutants in the bulk solution and subsequently enhance the pollutant removal efficiency [105,115,118,123–126]. In general, the use of NaCl as a supporting electrolyte was found to be better at accelerating the degradation rate of pollutants over other electrolytes [115,123,124]. For instance, the decolourisation of Alizarin Red S dyes was complete after 6 min of a sonoelectrochemical process using NaCl, while it required >120 min to obtain a similar removal level when Na_2SO_4 was used as the electrolyte [115]. This was attributed to the electrochemical production of active chlorine species (e.g. HClO , ClO^- , Cl_2) that can also oxidise pollutants through the processes described in Eqs. (7)–(12) (where “)))” indicates the application of ultrasound): [115,124,127–129]



Although persulfate and sulfate radicals can also be produced by oxidising sulfate-containing solutions, their mechanisms of oxidation are slower than chlorination [115,124]. The rapid oxidation mechanism of such active chlorine species converts organic pollutants into organochlorinated compounds, and this has raised some concerns since these organochlorine products may themselves constitute persistent organic pollutants which are more toxic than the parent molecules [115,123,124,130]. Hence, sonoelectrochemical research has mostly shifted to the use of sulfate-containing electrolytes, which are less likely to produce toxic by-products.

Apart from the type of electrolyte, the electrolyte concentration affects the degradation of pollutants by influencing the efficiency of both the ultrasonic and electrochemical processes. An increase of electrolyte concentration means enhanced ionic strength and solution conductivity, favouring faster degradation rates of pollutants [33,75]. In addition, the

presence of electrolyte alters the surface tension and vapour pressure of the solution, leading to more violent collapse of bubbles during cavitation, in turn leading to the production of more radicals [76,95,96,106]. Furthermore, the presence of inorganic salts in solution can result in a greater accumulation of the (generally more hydrophobic) organic pollutants at the bubble-bulk interface, resulting in more efficient degradation since the pollutants are closer to the cavitation event and the source of the radicals [95,106,110]. Previous studies showed that an increase in electrolyte concentration could improve the sonoelectrochemical treatment process through the synergistic benefits of sonolysis and electrolysis. For instance, the removal efficiency of ibuprofen by a sonoelectrochemical process improved from 58 to 77% when the electrolyte (Na_2SO_4) concentration was increased from 0.01 to 0.05 M [131].

The enhancement of degradation efficiency in most of these electrolyte effect studies is attributed to the production of more hydroxyl radicals as a result of more frequent and intense bubble collapse. However, it is also often the case that other radicals produced from the electrolytes are implicated in the mode of pollutant degradation. For example, Patidar et al. [93,94] exploited the anodic oxidation of sulfate ions (from a Na_2SO_4 -containing electrolyte), which produces the oxidising agent persulfate ($\text{S}_2\text{O}_8^{2-}$). Persulfate can then diffuse into the bulk and degrade the organic pollutants as part of a mediated homogeneous reaction [132,133]. However, there appears to be an optimal electrolyte concentration, above which the effectiveness of oxidation of pollutants is impaired [114]: Patidar et al. [94] found that once the concentration of Na_2SO_4 exceeded 1.5 g/L, the degradation of organic pollutants in their cosmetic wastewater treatment process started to decline. The authors claimed that at higher concentrations of electrolyte, sulfate and hydroxyl radicals might combine to produce persulfate, that in turn also contributes to the scavenging of hydroxyl radicals to produce other non-reactive compounds [93]. Similar trends were reported by Dominguez et al. [128] in that higher chloride concentration was held to promote the generation of substantial amounts of unwanted chlorine species, (i.e. ClO_2 , ClO_3^- and ClO_4^-), which consumed hydroxyl radicals to generate species with lower oxidising potentials, thus reducing the degradation efficiency.

The above reports make an interesting contrast with a series of studies conducted by the González-García group into sonoelectrochemical pollutant degradation in the absence of any electrolyte. Hence, Sáez et al. [134] carried out fixed current electrolytic degradation of perchloroethylene using Na_2SO_4 as a supporting electrolyte, and compared the effectiveness of this method to that of an electrolysis at fixed voltage in pure water. It was found that in the absence of background electrolyte, minor improvements were observed in the fractional conversion and degradation efficiency of perchloroethylene. The current efficiency when using a supporting electrolyte was around 4–5% regardless of concentration; however, when this electrolyte was removed the current efficiency increased to as much as 21%. Applying a comparable method to the high-frequency sonoelectrochemical degradation of trichloroacetic acid, the same group found similar benefits when operating in the absence of a supporting electrolyte [135]. After a 10-hour sonoelectrochemical treatment, degradation efficiencies of 55.3% and 43.2% were achieved at Na_2SO_4 concentrations of 10 and 50 mM respectively, whereas in the absence of Na_2SO_4 , a degradation efficiency of 67.2% was achieved. In a later publication under similar conditions, the same group was able to optimise their electrode configurations to achieve a degradation efficiency of ~ 97% using a Pb cathode and a Pt/Ti anode [136]. The authors proposed that their results could be developed into an environmentally-friendly method for water treatment that does not require the addition of extraneous reagents or secondary decontamination processes.

These findings indicate the importance of controlling the electrolyte concentration within a certain range in order to obtain optimal performance. Having said that, maintaining such a balance could well pose a challenge to the process operator as real-world industrial wastewaters

often contain insufficient or excessive concentrations of ions. Furthermore, the existence of multiple different ions in real-world waste streams might complicate the situation further due to the complex interplay between the various ions (see Section 3.5) [94].

2.9. Temperature

To date, the effect of temperature on the performance of sonoelectrochemical processes has not been thoroughly studied. The temperature of a solution has a significant effect on cavitation, with elevated temperatures disrupting solvent–solute interactions and facilitating higher mass transfer coefficients and kinetic parameters [137]. The general perception is that an increase in temperature will lead to a rise in the reaction rates of pollutants with reactive oxidising species [101,129]. Temperature also alters the viscosity and vapour pressure of the liquid which has a direct influence on the cavitation process [100,113]. Higher operating temperature increases the vapour pressure of the solution and reduces the viscosity and surface tension of the liquid, which leads to the easy generation of more cavitation bubbles [67,113]. However, the bubble collapse during cavitation is less intense and so is held to lead to lower hydroxyl radical production [67,135,137]. Also, the presence of excessive bubbles may disrupt the distribution of ultrasonic power into the entire solution. This might explain why inconsistent results have been reported on the influence of temperature on the sonoelectrochemical degradation of pollutants. For instance, Ren et al. [95] noticed that the degradation of chlorpyrifos at 30 °C was 28% higher than the rate at 5 °C. This improvement was attributed to increased formation of the oxidant $\text{S}_2\text{O}_8^{2-}$ at higher temperatures [138]. However, no significant differences in degradation efficiency were observed for the case of phenol or diuron within the temperature range of 15–35 and 10–40 °C, respectively [110,139,140]. On the other hand, Chen et al. [109] discovered that the oxidative destruction of nitrotoluene was favoured at lower temperatures within the operating range 30–45 °C, which the authors asserted was related to the solubility of oxygen in water, with higher oxygen solubility at lower temperatures allowing increased production of the oxidant hydrogen peroxide (from cathodic reduction of oxygen).

Determining the full effects of temperature on cavitation remain challenging, as this is often complicated by the influence of other competing/interdependent factors such as the number of bubbles produced, ultrasound intensity, etc. Findings from purely sonochemical studies have shown that an increase in liquid temperature will result in higher liquid vapour pressure, which in turn will trap more water vapour in the bubbles [141,142]. This can promote the production of free radicals from the dissociation of water vapour molecules and contribute to the increase in pollutant degradation rate. Furthermore, it is to be expected that temperature also will affect the stability and lifetime of the electrodes. For example, excessive temperatures could lead to the dissolution of catalytic layers on the electrode [143]. Unfortunately, such information on electrode stability at high temperature is generally lacking for sonoelectrochemical studies, which constitutes a further knowledge gap in terms of optimising the performance of sonoelectrochemical processes.

2.10. The specific nature of the pollutant

The characteristics of specific pollutants such as their hydrophilicity, solubility and volatility also affect the effectiveness of sonoelectrochemical processes in wastewater treatment. It is generally recognised that organic pollutants with high volatility will be incinerated inside the cavitation bubbles while hydrophilic or hydrophobic compounds with low volatility will be oxidised in bulk solution or at the bubble-solution interfaces by reaction with hydroxyl radicals ejected from the cavitation bubbles [142,144–146]. Hydrophobic pollutants with lower solubility and higher fugacity will accumulate at the bubble/solution interface and be degraded very effectively since the

concentration of hydroxyl radicals is relatively high at this interface [101,122,142]. On the other hand, hydrophilic pollutants tend to remain in bulk solution, and hence their degradation depends on the availability of free radicals (from cavitation bubbles or by direct generation at the electrodes) in bulk solution, with comparatively less degradation taking place at the bubble interface [101,122,135]. The pollutants can also sometimes exist in different forms according to the solution conditions (e.g. charged or neutral, or tautomers), which will impact their hydrophilicity and solubility in water, as well as the sonoelectrochemical degradation pathways and their efficiency (see example in Section 3.1) [101]. Understanding the nature of the target pollutants should enable some optimisation of the appropriate operating conditions for effective sonoelectrochemical degradation.

2.11. Energy consumption

The high energy consumption of sonoelectrochemical processes is one of the major obstacles hindering their wider application, despite often promising performance in degrading persistent organic pollutants [147]. Typically, in lab-scale studies, the energy consumption of a sonoelectrochemical process is expressed in the form of specific energy consumption (electrical energy consumed to degrade a certain amount of pollutant, or volume of solution) or energy efficiency (amount of pollutant degraded divided by the energy input required). A general rule of thumb is that the sonoelectrochemical process is more energy efficient when operated at higher electrolyte concentration, current density, and applied voltage [110]. For instance, the energy efficiency of a sonoelectrochemical system treating 400 mL of phenol solution (1 mM) with Na₂SO₄ electrolyte increased from 0.520 (±0.063) to 1.354 (±0.061) mmol/kWh when the electrolyte concentration was increased from 0.39 to 4.26 g/L [94].

The relationship between pollutant removal efficiency, speed of removal and process energy consumption can be complex, and can give rise to seemingly contradictory results. Hence Shestakova et al. [148] showed that the energy consumption required for 90% degradation of formic acid by a sonoelectrochemical process was 1785 kWh/m³, which was greater than the energy consumption of a standalone purely electrochemical process at 675 kWh/m³. In another case, Lima Leite et al. [68] observed that electro-oxidation alone required 54 min to degrade 32% of a sample of 2,4-dihydroxybenzoic acid, while it only needed 17 min to attain a similar level of degradation with the addition of 20 kHz ultrasound. Although the electrolysis time was reduced by a factor of 3, the much higher ultrasonic power consumption (23.4 W) as compared to that for the electrochemical-only route (1.75 W) resulted in a much higher energy consumption for the sonoelectrochemical process. On the other hand, there are also studies showing that the total energy consumption of a degradation process can be reduced in the presence of ultrasound. For instance, the energy consumption of the electrochemical and sonoelectrochemical processes for the degradation of Reactive Red 195 dye were 72.3 and 65.7 kWh/kg of chemical oxygen demand removal, respectively [129]. Another study showed that the total energy consumption for the degradation of Reactive Blue 19 dye decreased from 1.2 to 0.75 kWh/L when ultrasound of 80 kHz was applied to the electrochemical system [117].

In general, the ultrasonic system contributes significantly to the total energy consumption of a sonoelectrochemical process, and the ultrasound power requirement can be several orders of magnitude higher than that of the electrochemical system. This high energy consumption can be attributed to inefficient energy conversion, with only ~ 30% of the electrical energy being converted to cavitation energy [149]. Improvements to the efficiency of the ultrasound system are therefore of critical importance for driving down the energy consumption of sonoelectrochemical water treatment processes. One strategy that could be investigated in this regard is operating the ultrasound system in pulse mode (see Section 2.2). Operating in this fashion has been shown to cut the energy consumption for *p*-nitrophenol degradation from 0.7336

kWh/L (in continuous mode) to 0.3639 kWh/L (in pulse mode), whilst also improving the extent of degradation slightly (from 89% continuous to 94% pulsed) [76]. Similar improvement has also been reported by Xia et al. [75], where the energy consumption of a dual-pulse sonoelectrochemical process was 53.5% lower compared to a process operated with continuous ultrasound when degrading nitrobenzene. These findings indicate the potential to drastically decrease the energy consumption when the ultrasound system is operated creatively.

3. Recent examples of sonoelectrochemical wastewater treatment

The above discussion of the fundamentals of sonoelectrochemistry and the important factors that affect the operation of sonoelectrochemical processes for the degradation of organic pollutants sets the scene for an in-depth analysis of recent examples of the application of sonoelectrochemistry for wastewater treatment (mostly from 2015 onwards). In the following, sonoelectrochemical degradation treatments are classified by pollutant type, including dyes, pharmaceuticals, pesticides and surfactants, finishing with an overview of systems that aim to decontaminate real-world wastewater streams.

3.1. Dyes

The sonoelectrochemical decomposition of methylene blue [64,151], anthraquinone C.I. reactive blue 49 [107,122], naphthol blue black [150], acid orange 7 [91], and malachite green [33] have all been investigated in recent years; in many cases near-complete removal of dye was reported (see Table 1). In 2018, Zhu et al. [115] reported the degradation of Alizarin Red S using a boron doped diamond anode and a stainless steel cathode. Although a variety of conditions were tested, optimal results were achieved using an electrolyte consisting of 100 mg L⁻¹ of the dye and 0.05 M Na₂SO₄ at a pH of 4.97 and at 30 °C. Sonoelectrochemical oxidation (at a current density of 100 mA cm⁻², and 20 kHz (100 W power)) yielded complete discolouration of the solution, and > 86% chemical oxygen demand removal was achieved after 3 h. The authors used gas chromatography coupled to mass spectrometry (GC-MS) to identify some of the degradation products and assemble a possible degradation pathway (Fig. 5). The sample taken after 5 min and analysed by GC-MS indicated that the dye first loses its sulfonic acid group, forming either 1,2-dihydroxyanthraquinone or 9,10-anthracenedione. Measurements recorded after 30 min suggested that these intermediates were then broken down into a series of anhydrides and esters thought to contain phthalic anhydride, phthalic acid diesters, and methyl 4,4-dicaprylate. After 180 min of the sonoelectrochemical oxidation treatment, the main degradation product present was identified as bisphenol A, which can be mineralised to CO₂ and H₂O. This finding differed from previously reported degradation pathways of Alizarin Red S by purely electrochemical oxidation, in which the main intermediates were found to be phthalic acid and small molecule carbonyl compounds which were then mineralised [160,161].

In a study published in 2020, Ghanbari et al. [91] coupled ultrasound to the electroperoxone method for the degradation of the dye acid orange 7 (AO7). The electroperoxone method refers to the in-situ electrochemical generation of peroxone (i.e. H₂O₂ and O₃), leading to the production of hydroxyl radicals. In short, oxygen is reduced at the cathode to H₂O₂, which may then react with ozone bubbled into solution, forming hydroxyl radicals [80,162]. Additionally, hydroxyl radicals can be formed through the reduction of ozone (Eqs. (13)–(15)): [163,164]

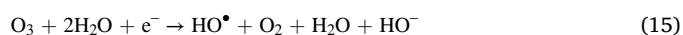
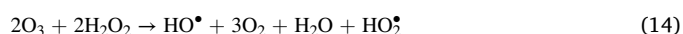
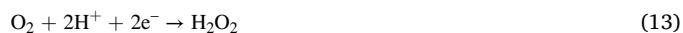


Table 1

A summary of the reports of the sonoelectrochemical degradation of organic pollutants in water (since 2015). EC = electrochemical; US = ultrasound; TOC = total organic carbon; COD = chemical oxygen demand.

Entry	Pollutant	Initial Pollutant Concentration	Electrodes	Electrolyte	Experimental Conditions	Efficiency	Reference
1	Methylene Blue	0.025 mM (~8 mg L ⁻¹)	Anode: Ti/Ta ₂ O ₅ -SnO ₂ Cathode: Ti plate	0.1 M Na ₂ SO ₄	EC – 20 mA (9.09 mA cm ⁻²), US – 850 kHz (0.186 Wcm ⁻³)	Complete decolourisation of dye solution after 45 mins. COD reduction of 85.4% after 2 hrs.	Shestakova et al. [64]
2	Anthroquinone C.I. Reactive Blue 49	100 mg L ⁻¹	Anode: Carbon Cathode: Pt Plate	10 M H ₂ O ₂ , H ₃ PO ₄ + NaH ₂ PO ₄ buffer	EC – 0.7 V, US – 35 kHz (150 Wcm ⁻²)	99% decolourisation was observed along with 87.8% COD reduction.	Radi et al. [122]
3	Anthroquinone C.I. Reactive Blue 49	100 mg L ⁻¹	Anode: Carbon Cathode: Pt Plate	H ₃ PO ₄ + NaH ₂ PO ₄ buffer	EC – 0.7 V, US – 35 kHz (150 Wcm ⁻²)	After 80 mins, 97.2% decolourisation and 91% COD removal.	Radi et al. [107]
4	Alizarin Red S	100 mg L ⁻¹	Anode: BDD Cathode: Stainless Steel	0.05 M Na ₂ SO ₄	EC – 100 mA cm ⁻² , US – 20 kHz (100 W)	Complete decolourisation of solution, and 86.07% COD removal after 3 hrs.	Zhu et al. [115]
5	Napthol Blue-Black	3.2 μM (~1.97 mg L ⁻¹)	Anode: BDD Cathode: Graphite	0.05 M (NH ₄) ₂ SO ₄ / 1 M H ₂ SO ₄	EC – 23 mA cm ⁻² , US – 37 kHz	Extent of mineralisation not determined.	Wallace et al. [150]
6	Acid Orange 7	50 mg L ⁻¹	Anode: Pt Sheet Cathode: Graphite Felt	10 mM Na ₂ SO ₄	EC – 300 mA (33.3 mA cm ⁻²), US – 20 kHz (100 W) Ozone dosage – 33 mg/L	Complete decolourisation of solution, and 88% COD removal after 1 hr.	Ghanbari et al. [91]
7	Methylene Blue	14.21 mg L ⁻¹	Anode: Zn ₃ (PO ₄) ₂ Cathode: Pt	0.1 M NaCl	EC - N.G., US – 40 kHz (180 W)	After 60 mins, the concentration of dye is reduced by 91 %.	Chennah et al. [151]
8	Allura Red	100 mg L ⁻¹	Anode: Ti-PbO ₂ Cathode: Stainless Steel	0.05 M Na ₂ SO ₄	EC – 0.75 A (18.5 mA cm ⁻²), US – 40 kHz	Complete removal of dye after 60 mins. Sonoelectrochemical method showed a 9% increase in COD removal versus the non-sonicated method.	Kacem et al. [105]
9	Erythrosine B	100 mg L ⁻¹	Anode: Ti-PbO ₂ Cathode: Stainless Steel	0.05 M Na ₂ SO ₄	EC – 0.75 A (18.5 mA cm ⁻²), US – 40 kHz	Complete removal of dye after 90 mins. Sonoelectrochemical method showed an 11% increase in COD removal versus the non-sonicated method.	Kacem et al. [105]
10	Malachite Green	100 mg L ⁻¹	Anode: Stainless steel, titanium, aluminum, Ru-Ir coated titanium, Ru-Ta coated titanium, graphite, brass and copper. Cathode: Graphite	15 g/L Na ₂ SO ₄	EC – 20 V, US – 25 kHz (300 W)	After 60 mins, the dye was degraded by 94.92% when using Ru-Ir coated Ti working electrode.	Ren et al. [33]
11	Carbamazepine	10 mg L ⁻¹	Anode: Ti-PbO ₂ cylinder Cathode: Ti cylinder	Wastewater effluent	EC – 4.86 A (~11.6 mA cm ⁻²), US – 520 kHz (38.29 W)	Carbamazepine concentration had decreased by 93% after 177 mins. TOC and COD removal was 60% and 93%, respectively.	Tran et al. [152]
12	Diclofenac	50 μg L ⁻¹	Anode: BDD Cathode: BDD	30 mM Na ₂ SO ₄	EC – 7.2 V, US – 850 kHz (120 W)	After 5 mins, concentration of diclofenac had reduced by > 90%.	Finkbeiner et al. [86]
13	p-Nitrophenol	100 mg L ⁻¹	Anode: Ti/Sb-SnO ₂ Cathode: Ti	0.05 M Na ₂ SO ₄	EC – 10 V, US – 850 kHz (120 W)	After 2 h of alternating electrode potential and ultrasonic pulses, 94.1% of p-nitrophenol was degraded.	Xie et al. [76]
14	Cephalosporin Wastewater	COD – 326.9 mg L ⁻¹	Anode: Ru/Ir nanocoated electrode Cathode: Stainless Steel	Wastewater effluent	EC – 8 mA cm ⁻² , US – 45 kHz (100 W)	94% removal efficiency for COD removal after 30 mins.	Yang et al. [40]
	Chlortetracycline	10 mg L ⁻¹					

(continued on next page)

Table 1 (continued)

Entry	Pollutant	Initial Pollutant Concentration	Electrodes	Electrolyte	Experimental Conditions	Efficiency	Reference
15			Anode: Ti-PbO ₂ Cathode: Ti	Wastewater effluent	<i>EC</i> – 3.92 A (14 mA cm ⁻²), <i>US</i> – 500 kHz (20 W) <i>EC</i> – 4.86 A, <i>US</i> – 520 kHz (38 W)	Chlortetracycline concentration was reduced by 98% after 95 mins.	Tran et al. [153]
16	Carbamazepine	10 mg L ⁻¹	Anode: Ti-PbO ₂ Cathode: Ti	0.01 M Na ₂ SO ₄	<i>EC</i> – 4.86 A, <i>US</i> – 520 kHz (38 W)	86% reduction in carbamazepine, and 80.59% mineralisation after 180 mins.	Tran et al. [21]
17	Sulfamethoxazole	20 mg L ⁻¹	Anode: Pt Cathode: Graphite	0.1 M Na ₂ SO ₄	<i>EC</i> – 20 mA cm ⁻² , <i>US</i> – 40 kHz (100 W)	Sulfamethoxazole concentration decreased by 83% after 60 mins.	Huang et al. [106]
18	Bisphenol A	1 mg L ⁻¹	Anode: BDD Cathode: BDD	30 mM Na ₂ SO ₄	<i>EC</i> – 20 mA cm ⁻² , <i>US</i> – 24 kHz (W)	>90% removal of bisphenol A after 30 mins.	Dietrich et al. [63]
19	Ibuprofen	5 mg L ⁻¹	Anode: Pt Cathode: Carbon cloth coated with Carbon black.	0.05 M Na ₂ SO ₄	<i>EC</i> – 0.03 A, <i>US</i> – 37 kHz (320 W)	84.2% removal after 60 mins using an oxygen-permeable cathode.	Darvishi Cheshmeh Soltani et al. [131]
20	Atrazine	TOC – 40.89 g L ⁻¹	Anode: BDD Cathode: Steel	Wastewater effluent	<i>EC</i> – 8 mA cm ⁻² , <i>US</i> – 24 kHz (200 W)	80% TOC removal after 30 mins, 100% after 250 min.	dos Santos et al. [154]
21	Piroxicam	245 µg L ⁻¹	Anode: BDD Cathode: Stainless Steel	0.1 M Na ₂ SO ₄	<i>EC</i> – 13 mA cm ⁻² , <i>US</i> – 20 kHz (14 W/L)	Complete degradation of piroxicam in < 10 mins.	Kouskouki et al. [125]
22	Ofloxacin	2 g L ⁻¹	Anode: TiRuO ₂ Cathode: Steel	2.0 g L ⁻¹ Na ₂ SO ₄	<i>EC</i> – 214 A cm ⁻² , <i>US</i> – 33 kHz (54 W)	91.2% ofloxacin removal, 70.12% COD removal after 120 mins.	Patidar et al. [93]
23	E.Coli	16,000 CFU 100 mL ⁻¹	Anode: BDD Cathode: Stainless Steel	Wastewater effluent	<i>EC</i> – 1.27 A cm ⁻² , <i>US</i> – 200 W	Complete disinfection in ~ 60 mins.	Llanos et al. [155]
24	Trichloroacetic Acid	0.5 mM (81.69 mg L ⁻¹)	Anode: Pt/Ti Cathode: Ti, AISI 304 stainless steel, carbon and Pb	Water	<i>EC</i> – 4 mA cm ⁻² or 14.3 V <i>US</i> – 863 kHz (0.054 W cm ⁻³) <i>EC</i> – 83.64 mA cm ⁻² ,	Using Pb cathode, a degradation efficiency of 96.9% was achieved after 600 mins.	Esclapez et al. [136]
25	Perfluorooctanoic Acid	94 mM (38.92 g L ⁻¹)	Anode: Ti/PbO ₂ Cathode: Stainless Steel	75 mM Na ₂ SO ₄	<i>EC</i> – 83.64 mA cm ⁻² , <i>US</i> – 20 kHz	95.48% mineralisation after 90 mins.	Bonyadinejad et al. [156]
26	Methyl Paraben	100 mg L ⁻¹	Anode: BDD Cathode: Stainless Steel	3.0 g L ⁻¹ Na ₂ SO ₄	<i>EC</i> – 30 mA cm ⁻² , <i>US</i> – 20 kHz (75% power, instrument had a nominal power of 200 W)	Complete mineralisation of methyl paraben in < 5 h.	Dionisio et al. [130]
27	Thiram	100 mg L ⁻¹	Anode: BDD Cathode: Pt Net	0.05 M K ₂ SO ₄ 30% MeOH	<i>EC</i> – 50 mA cm ⁻² , <i>US</i> – 20 kHz (523 W cm ⁻²)	100% removal of the pollutant after 2 h.	Steter et al. [103]
28	Formic Acid	250 mg L ⁻¹	Anode: Ti/Ta ₂ O ₅ -SnO ₂ Cathode: Ti plate	3.0 g L ⁻¹ NaCl	<i>EC</i> – 20 mA (~9.1 mA cm ⁻²), <i>US</i> – 381 kHz (0.02 W cm ⁻³)	Over 99% degradation of formic acid after 2 h.	Shestakova et al. [148]
29	Pulp and effluent	4 mg L ⁻¹	Anode: Fe Cathode: Fe	Wastewater effluent	<i>EC</i> – 4 A dm ⁻² , <i>US</i> – 40 kHz (350 W)	After 4 h, 100% decolourisation and 95% COD removal.	Asathambi et al. [157]
30	Microcystin-LR	10 µg L ⁻¹	Anode: BDD Cathode: Stainless Steel	Wastewater effluent	<i>EC</i> – 6 mA cm ⁻² , <i>US</i> – 20 kHz (15 W)	93% degradation when process was carried out over 5 mins; 99% degradation was achieved after 10 mins.	Gao et al. [20]
31	Chlorpyrifos	900 mg L ⁻¹	Anode: Stainless Steel	2.0 g L ⁻¹ Na ₂ SO ₄	<i>EC</i> – 20 V, <i>US</i> – 40 kHz (200 W)	After 60 mins, 93.3% degradation of chlorpyrifos was observed.	Ren et al. [95]

(continued on next page)

Table 1 (continued)

Entry	Pollutant	Initial Pollutant Concentration	Electrodes	Electrolyte	Experimental Conditions	Efficiency	Reference
32	Methyl Paraben in Synthetic Micellar	DOC – 400 mg L ⁻¹	Cathode: Stainless Steel Anode: BDD	Synthetic Wastewater	EC – 30 mA cm ⁻² , US – 20 kHz	90% mineralisation was achieved in a reaction time of 8 h.	Dionisio et al. [124]
33	Wastewater Propyl Paraben	TOC – 7.7 mg L ⁻¹	Cathode: Stainless Steel Anode: BDD Cathode: Ti Sheet	0.1 M Na ₂ SO ₄	EC – 3.785 mA cm ⁻² , US – 20 kHz (36 W L ⁻¹)	TOC removal of 36% after 15 mins.	Frontistis [158]
34	Perfluorooctanoic Acid	60 mg L ⁻¹	Anode: Ti/SnO ₂ -Sb/Ce-PbO ₂	50 mM Na ₂ SO ₄	EC – 5 V, US – 20 kHz (52 W)	98.7 % of the contaminant was removed after 180 mins.	Xu et al. [42]
35	4-Chlorophenol	COD – 338 mg L ⁻¹	Cathode: Ti Sheet Anode: Titanium suboxide-REM	50 mM NaClO ₄	EC – 3 V, US – 28 kHz (180 W)	After 100 successive cycles of 3 V for 40 mins, 87.81%-88.95% COD removal was maintained.	Teng et al. [159]
36	Wastewater Sludge	SCOD –37.6 mg L ⁻¹	Cathode: Stainless Steel Mesh Anode: Ti/PbO ₂ -Fe-SS-SS electrode system	Wastewater effluent	EC – 100 A cm ⁻² , US – 40 kHz (180 W)	After 30 mins, a degree of degradation of 25.3% was achieved.	Ozyonar et al. [85]
37	Cosmetic Industry Wastewater	COD – 2350 mg L ⁻¹	Anode: Ti/RuO ₂ Cathode: Stainless Steel	Wastewater effluent	EC – 213 A cm ⁻² , US – 33 kHz (100 W)	80.9% COD removal efficiency after 180 mins.	Patidar et al. [94]

The application of ultrasound to this process would be expected to further increase radical production, and indeed, complete discolouration and 88% mineralisation were achieved using a current of 300 mA (Pt sheet anode, graphite cathode), with sonication at 20 kHz (100 W), and an ozone dosage of 33 mL min⁻¹, at pH = 7 for 1 h. This process was benchmarked against several other oxidation techniques, including the purely electroperoxone method, sonication alone, ozone oxidation, and a combination of ultrasound and ozone oxidation: it was found that the ultrasound-assisted electroperoxone method provided the highest degradation rate. This study also reported a strong synergistic effect, meaning the degradation rate achieved in the combined method was greater than the sum of the results from the electrochemical and sonication techniques. The synergistic index (SI), given by Eq. (16), was found to be equal to 1.95.

$$SI = \frac{k_{\text{electroperoxone+sonication}}}{k_{\text{electroperoxone}} + k_{\text{sonication}}} \quad (16)$$

In both examples highlighted in this section thus far, the chosen anode materials were boron doped diamond and Pt respectively. Though undoubtedly effective for use in pollutant degradation, these materials are costly and could impede the scaling-up of such processes. Thus, recent efforts have focussed on using cheaper alternative electrodes for sonoelectrochemical dye removal. To this end, the sonoelectrochemical degradation of Allura Red AC and Erythrosine B, two food dyes, was of late investigated using a relatively low-cost Ti-PbO₂ anode. Using comparatively mild conditions (0.75 A, 40 kHz), Kacem et al. [105] achieved complete removal of Allura Red AC in 60 mins, and Erythrosine B in 90 mins. The time required to degrade the dyes was unchanged by the presence of ultrasound; however, the chemical oxygen demand removal was markedly improved by sonication (+9% for Allura Red AC, +11% for Erythrosine B) suggesting that sonication promotes complete mineralisation. Moreover, the effect of the supporting electrolyte was also established by running comparative tests with Na₂SO₄, NaCl, and Na₂CO₃; it was found that NaCl yielded the fastest degradations of each dye due to the generation of reactive chlorine species (Eqs. (7)–(12)).

Significant degradation of the dyes was also achieved when using Na₂SO₄ as an electrolyte due to the known ultrasound enhanced electro-

generation of sulfate radicals, which had previously been exploited by Dionisio et al. [130] for removal of the preservative methyl paraben from aqueous solution. More recently, a number of different anode materials were trialled for their suitability for the sonoelectrochemical degradation of malachite green [33]. Graphite, titanium, stainless steel, brass, copper, aluminium, iridium-tantalum-coated titanium, and ruthenium-iridium-coated titanium electrodes were all tested, using Na₂SO₄ as a supporting electrolyte over 60 mins at an applied voltage of 20 V and sonication at 25 kHz (300 W). Considering only the materials which achieved > 90% degradation of malachite green: copper and brass electrodes reacted with SO₄²⁻ ions in solution, forming copper sulfate, and thus were ill-suited over extended operation; graphite was found to corrode and fragment over the course of the experiment, necessitating further treatment before absorbance of the electrolyte could be measured; the stability of the aluminium anode was poor, and an oxidised film would form, and finally Ru-Ir coated Ti gave good performance, with no visible corrosion, thus making it the obvious choice for further experimentation. Under optimal conditions (see Table 1), this electrode material gave a malachite degradation efficiency approaching 95%. The authors then proposed a degradation pathway based on the identification of some intermediates; they postulated that HO₂ ions generated at the cathode attack both tautomeric forms of the malachite green cation, as shown in Fig. 6. A series of N-demethylation reactions then occur (disrupting the electron-donating properties of the nitrogen centres) before the resulting products are further oxidised.

3.2. Pharmaceuticals

Pharmaceuticals constitute a large and growing source of wastewater pollution. For example, somewhere between 50 and 90% of administered antibiotics and their primary metabolites are excreted [165]. This has fuelled research into developing new ways to remove such pollutants from the watercourse.

In 2017, Huang et al. [106] studied the sonoelectrochemical degradation of the antibiotic sulfamethoxazole. Using Pt and graphite as the anode and cathode respectively in 0.1 M Na₂SO₄ at a current density of 20 mA cm⁻² under sonication at 40 kHz (100 W), an 83% degradation

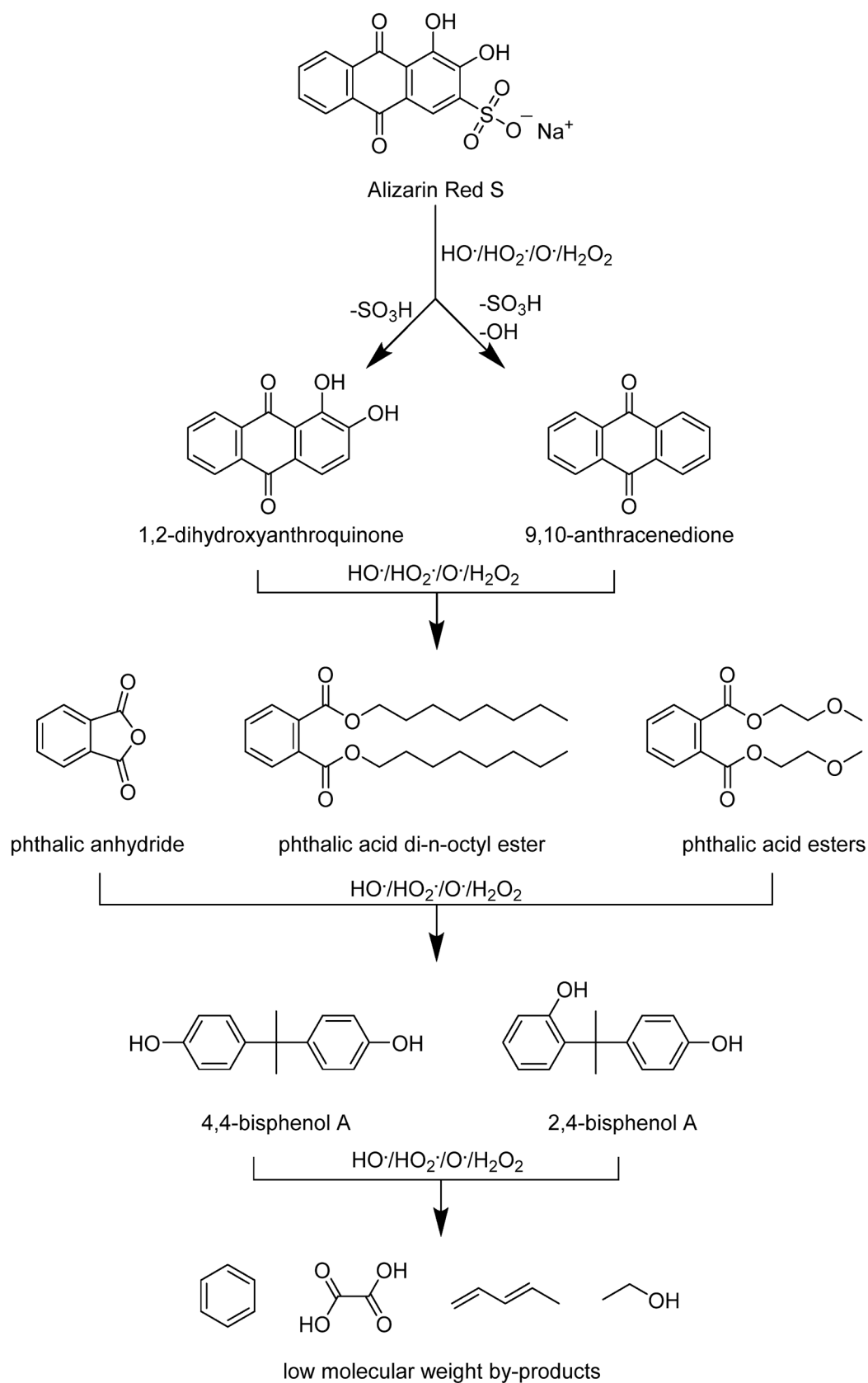


Fig. 5. The possible sonoelectrochemical degradation pathway of Alizarin Red S, as proposed by Zhu et al. [115].

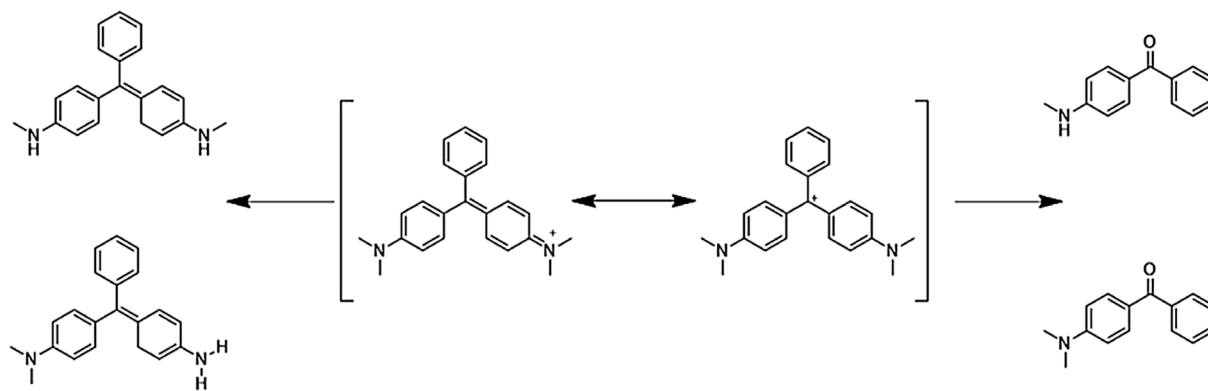


Fig. 6. The degradation products of malachite green identified by Ren et al. [33], formed via reaction of HO_2^- with the positively-charged imine ion (left) and the carbocation (right).

efficiency of sulfamethoxazole was observed after 60 mins. In comparison, under the same electrochemical conditions but in the absence of ultrasound, only 10% of the sulfamethoxazole was degraded, and using ultrasound alone resulted in only marginal degradation. Thus, a large synergy factor was evident in the combined sonoelectrochemical approach. A series of mechanistic studies were then performed to establish the primary oxidising species. The proposed mechanism (Fig. 7) suggests that oxygen is reduced at the graphite cathode, forming superoxide (O_2^-), which is reduced to H_2O_2 and then to HO^\bullet . The concurrent anodic reaction is the oxidation of H_2O and HO^- , ultimately leading to the production of HO^\bullet . Hydroxyl radicals are also produced as a result of acoustic cavitation events, as discussed previously, with the sulfamethoxazole being oxidised both directly at the anode and indirectly through these radicals.

The authors used high-performance liquid chromatography coupled with mass spectrometry to identify the organic intermediates hydroxylated sulfamethoxazole, 5-methyl-1,2-oxazol-3-amine, and 4-amino-hydroxybenzenesulfonic acid [106]. Using this information and examples in the literature, a possible degradation pathway was proposed (Fig. 8). To assist in de-convoluting the degradation pathway (both in the presence and absence of ultrasound), the authors employed the same method, but using NaCl as a supporting electrolyte. The researchers determined that, in both methods, two degradation mechanisms would occur due to oxidation from either HOCl or HO^\bullet . The hydroxyl radical-mediated degradation of sulfamethoxazole was found to be similar in the two experiments; however, fascinating insight was gained by comparing this with the HOCl pathway. In the absence of ultrasound, the electrooxidation of the pollutant was held to lead to the generation of

complex chlorinated intermediates, some of which would couple, resulting in a number of potentially harmful dimeric and polymeric chlorinated by-products. The HOCl -mediated oxidation of sulfamethoxazole in the sonoelectrochemical system was far simpler, with only two chlorinated products detected in the degradation pathway. It can be deduced from this experiment that, by using a combined sonoelectrochemical approach, the generation of harmful chlorinated by-products can be mitigated to some degree, a finding which could be important in the context of developing ultrasound-assisted electrochemical degradation processes in the presence of chloride ions (see also Section 2.8) [40,105,153,155].

The degradation of the anti-inflammatory drug ibuprofen was investigated by Soltani and Mashayekhi [131] using a sonoelectrochemical approach, employing a novel oxygen-permeable cathode: carbon black-coated carbon cloth. This group had previously demonstrated that the use of gas diffusion cathodes greatly promotes the electrochemical generation of H_2O_2 , suggesting that such cathodes would be effective in the removal of pollutants from aqueous solution [166]. A comparative degradation study of ibuprofen degradation (at a concentration of 5 mg L^{-1}) was performed over 60 mins, using a current of 0.3 A and 37 kHz sonication (320 W), in 0.05 M Na_2SO_4 . Optimal ibuprofen removal was achieved when using a combined sonoelectrochemical method; this study also confirmed that the use of the novel oxygen-permeable cathode resulted in a significant increase in removal efficiency (84%) compared to that achieved with a standard carbon cloth (73%). Operating the ultrasonic bath in a pulsed mode resulted in a further increase in removal efficiency, up to nearly 89%. Moreover, several degradation intermediates were identified using GC-MS,

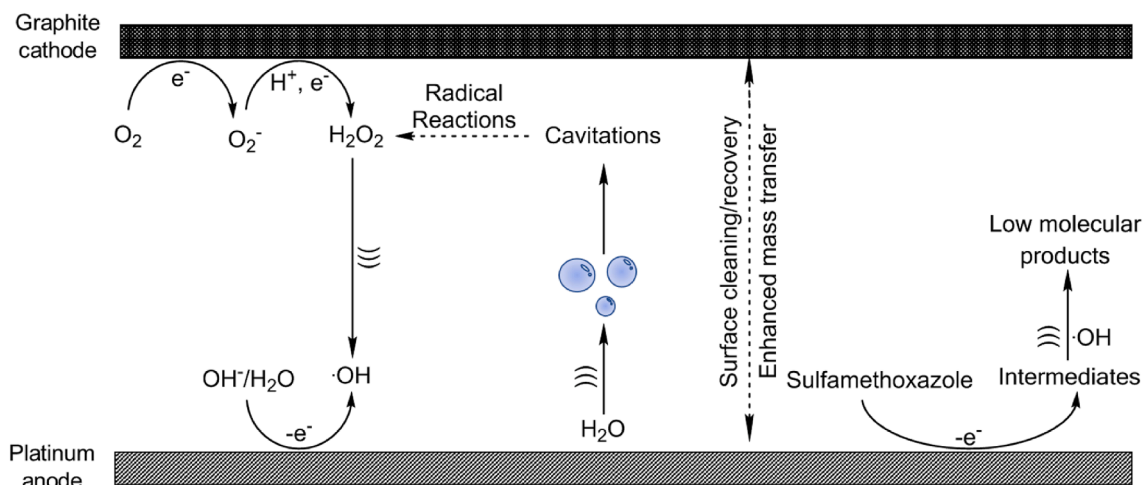
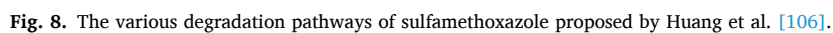


Fig. 7. The proposed mechanism of radical generation in the degradation of sulfamethoxazole proposed by Huang et al. [106].



suggesting a decomposition pathway of ibuprofen first to 1-(1-hydroxyethyl)-4-isobutylbenzene, which is then further oxidised to 4'-(2-methylpropyl)-acetophenone and 4-(2-methylpropyl)-phenol. The identification of 1-ethyl-4-isobutylbenzene indicated that a decarboxylation of ibuprofen also occurs (Fig. 9).

Oxofloxacin is an effective antibiotic, which, like other antibiotics, is bio-recalcitrant, with the potential to accumulate in wastewater as a pharmaceutical pollutant [167]. In light of this, Patidar and Srivastava [93] studied its electrochemical degradation using a two-electrode reactor (TiRuO₂ anode, steel cathode) fitted with an ultrasonic transducer (see also Section 2.7). Under optimal conditions (see Table 1), this method was able to effectively eliminate oxofloxacin (>91% oxofloxacin removed, 70% chemical oxygen demand removal), and again a synergistic relationship was confirmed in the combined sonoelectrochemical approach. The authors employed radical scavengers to confirm the role of radicals in this oxidation process; perhaps unsurprisingly, it was discovered that the hydroxyl radical was the most significant contributor. A variety of mass spectrometry-based techniques were used to identify intermediates and to propose a number of degradation pathways, as shown in Fig. 10.

3.3. Pesticides

Since the mid-20th century there has been a massive increase in the adoption of pesticides in order to boost agricultural productivity [168–171]. With current projections estimating that the global population may reach nearly 10 billion by 2050, it is likely that the use of these agents will only increase; however, many such pesticides can have a deleterious effect on the environment [170,172,173]. It is against this backdrop that sonoelectrochemistry was utilised by Esclapez et al. [136] to degrade the herbicide trichloroacetic acid. Noting that the primary electrochemical degradation pathway for trichloroacetic acid consists of repeated cleavage of the C–Cl bonds at the cathode, four different cathode materials were evaluated (Ti, AISI 304 stainless steel, carbon and Pb), and in each case the application of ultrasound was found to enhance the electrochemical degradation efficiency of trichloroacetic

acid. The Pb cathode yielded the best results, with almost complete dechlorination of trichloroacetic acid achieved after 600 min, and further tests suggested close to complete mineralisation of the acid. Although the Pb cathode was found to be the most effective for trichloroacetic acid degradation, the use of lead for water treatment processes is not ideal (see also Section 2.1) [174], especially in the context of high frequency ultrasound, as acoustic cavitation can erode the cathode causing Pb to leach into the “treated” water. It was found that the Pb content in the water after this sonoelectrochemical process (2.1 ppm) far exceeded the maximum value permitted according to the U.S. Environmental Protection Agency (0.015 ppm). The authors argued that a mitigating factor could be that the eroded Pb particles were also found to degrade trichloroacetic acid and in turn they could be electrodeposited at the anode as PbO₂, removing them from the water. However, the requirements to meet such low Pb levels in the treated water stream will doubtless render many way of developing sonoelectrochemical systems that are dependent on the use of lead.

Thiram is the second most widely-used contact herbicide in the world; [175] however, it can be incredibly harmful if ingested and is a known neurotoxin [176]. In 2016, it was demonstrated that thiram could be effectively degraded using a sonoelectrochemical approach with a boron doped diamond anode; under optimal conditions (see Table 1), complete degradation of thiram was achieved after 2 h [103]. A comparative study of the electro-oxidation of thiram (both in the presence and absence of ultrasound) allowed the researchers to propose decomposition schemes under each regime (Fig. 11). Common intermediates were identified in both pathways, such as the products formed through oxidation of the C=S groups in the thiram molecule, although it should be noted that several additional intermediates resulting from hydroxyl radical reactions were identified in the sonoelectrochemical reaction (blue dashed boxes in Fig. 11). In both approaches, the authors noted that the absence of lower molecular weight acids such as fumaric or oxalic acid was an indication that complete mineralisation of thiram was not achieved.

The removal of the pesticide chlorpyrifos via sonoelectrochemical oxidation using two stainless steel electrodes was reported in 2019 (see

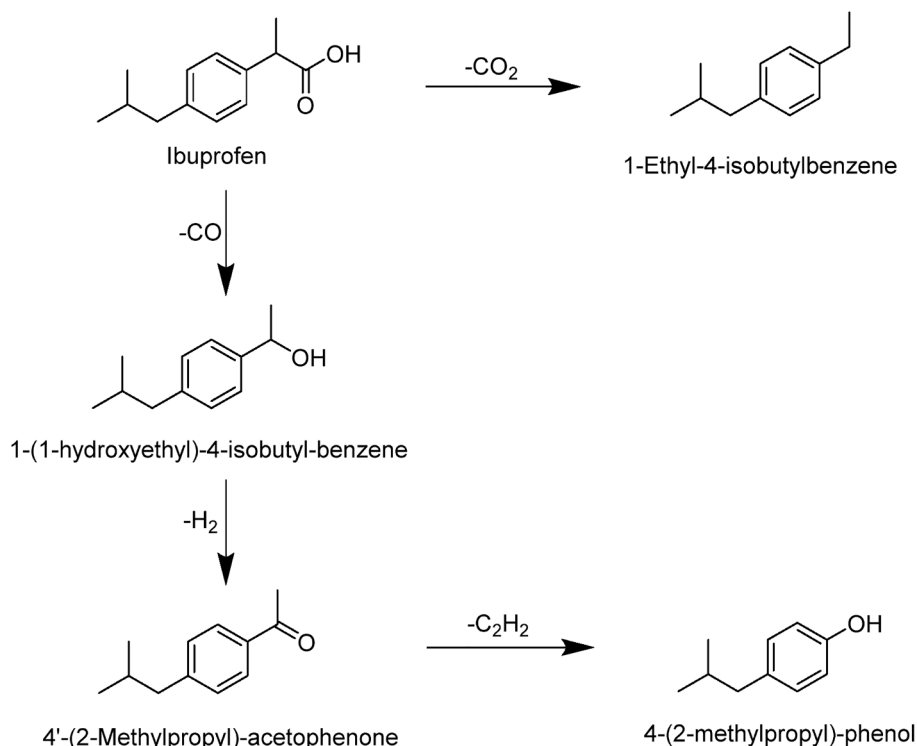


Fig. 9. The sonoelectrochemical degradation pathways of ibuprofen proposed by Soltani and Mashayekhi [131].

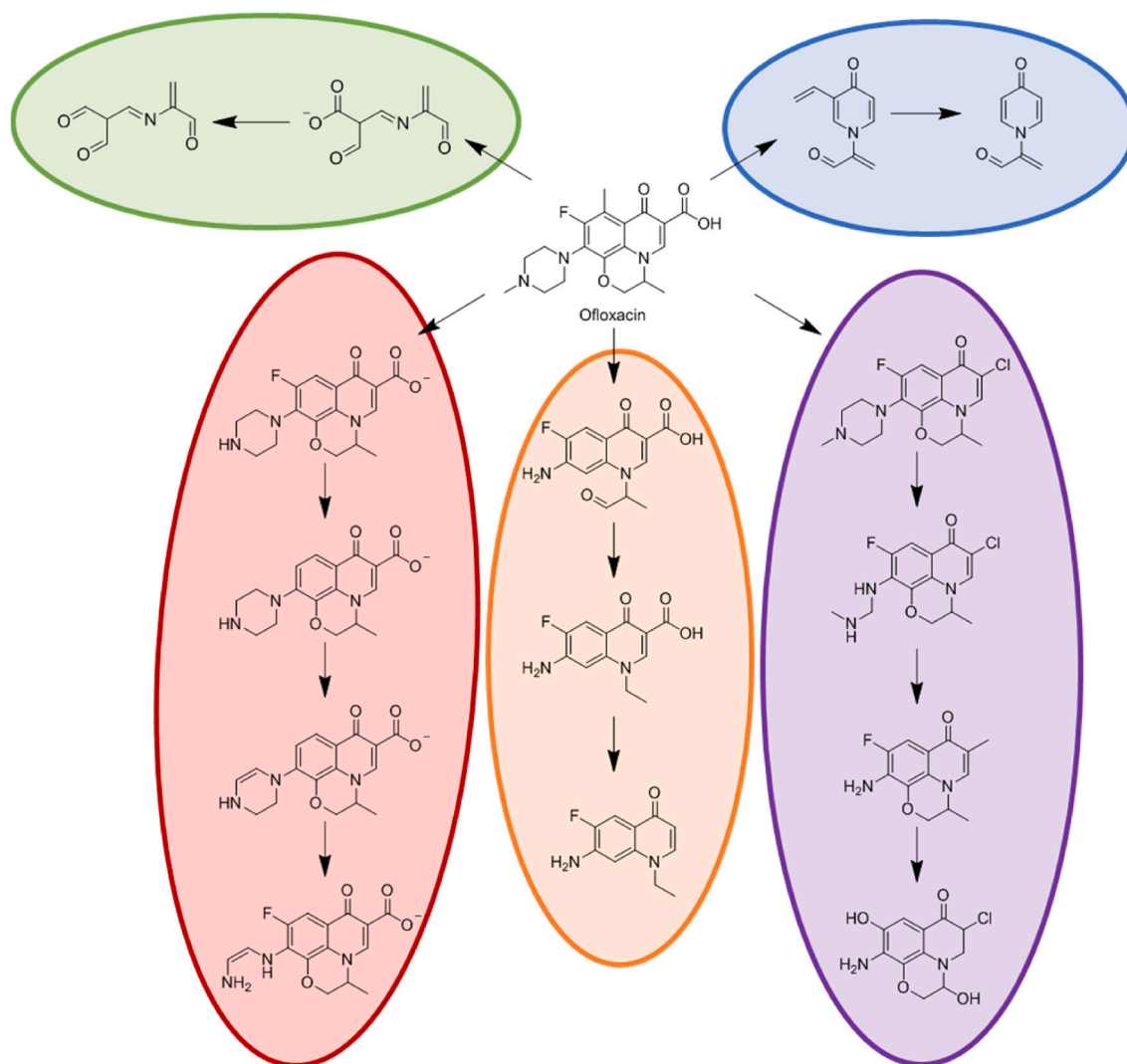


Fig. 10. The degradation pathways of ofloxacin proposed by Patidar and Srivastava [93].

also Section 2.9) [95]. Using a relatively high concentration of the pesticide (900 mg L^{-1}), a degradation of $> 90\%$ was achieved after 1 h (20 V, 40 kHz, 200 W); a synergistic factor between the electrochemical and ultrasonic inputs of 37% was also reported.

3.4. Surfactants

Surfactants are a class of molecules typified by being amphiphilic, generally with a hydrophobic tail and hydrophilic head [177,178]. Above a certain concentration they aggregate, forming micelles in which the surfactant heads point outwards creating a hydrophilic exterior which shields a hydrophobic pocket. The benefit of this is that micelles can entrap otherwise poorly soluble organic compounds, and greatly enhance their solubility [179]. However, the shielding effect of these micelles has been shown to impede the oxidation of target molecules [180–182], and so a treatment method which can be effective in the presence of micelles is of great interest.

Building on their previous work utilising electrochemical and sonoelectrochemical methods for the degradation of methyl paraben [130,183,184], Dionisio et al. [124] investigated the effectiveness of these techniques on the treatment of a synthetic micellar-containing wastewater, with high concentrations of organic matter, mimicking the composition of wastes produced in the cosmetic industry. To understand the behaviour of the micelles under the action of an ultrasonic

field, the researchers monitored particle size during electrolysis both in the presence and absence of sonication. Although it was anticipated that sonication would promote micelle degradation (as had previously been reported [185]) an initial growth in particle size was first observed under all sonication conditions. This growth was ascribed to incorporation into the micelles of gases produced during electrolysis, promoted by ultrasonic rupturing of the electro-generated bubbles. This initial growth was followed by a sharp decrease in size, suggesting that larger micelles are unstable and ultimately collapse, releasing their entrapped organic compounds into the bulk of the solution. Moreover, the extent of the initial growth was greatly dependent on the frequency of sonication. It was observed that the micellar growth was significantly higher when operating at a lower frequency (20 kHz) than at higher frequencies (1 or 10 MHz), leading the authors to speculate that the growth of the bubbles through the incorporation of gases was inhibited at higher frequencies.

The authors also reported that the application of ultrasound was effective in addressing physical features of wastewater which can negatively impact the efficiency of this process, finding that a faster depletion of turbidity and dissolution of foam was achieved. Despite this, when looking solely at the concentration of methyl paraben in the synthetic wastewater, no significant difference in its degradation was found in the presence or absence of ultrasound.

The degradation of the surfactant perfluorooctanoic acid has also been studied in some detail. In 2015, Bonyadinejad et al. [156] used a

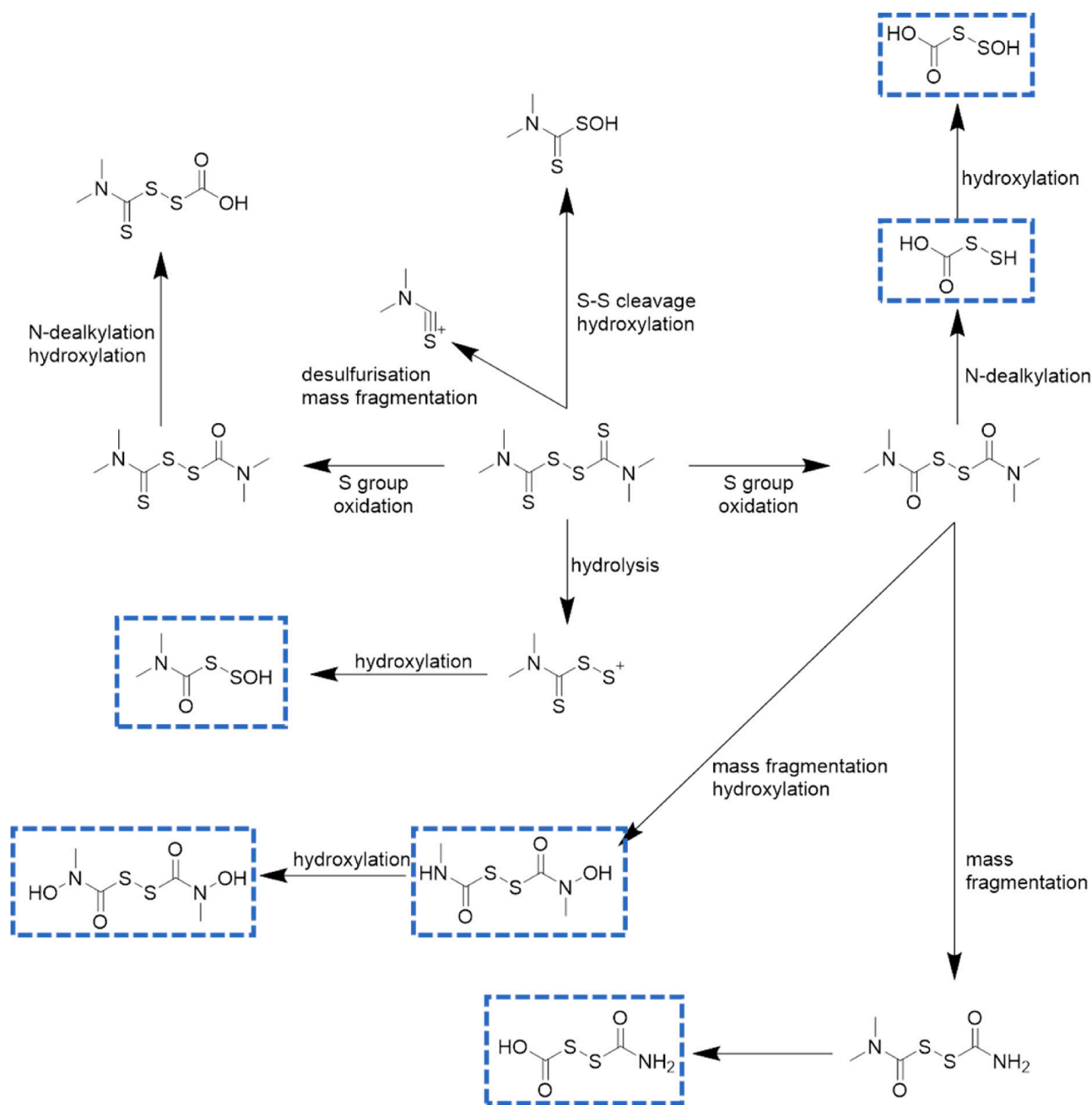


Fig. 11. The pathways of thiram degradation proposed by Steter et al. [103]. Structures without blue dashed boxes were found to be common to both the electrochemical oxidation and sonoelectrochemical oxidation regimes. Structures inside blue dashed boxes were only found in the sonoelectrochemical treatment route.

low-cost electrode setup (Ti/PbO₂ anode, Ti plate cathode) to effectively de-fluorinate perfluorooctanoic acid, achieving > 95% mineralisation in 90 mins under optimal conditions (see Table 1). A synergism was observed in the combined sonoelectrochemical method, with three possible explanations: (1) ultrasound promoted the formation of sulfate radicals in the supporting electrolyte, Na₂SO₄, (2) the mass transfer enhancements brought about by sonication increased diffusion of the produced sulfate/hydroxyl radicals at the electrode surface, and/or (3) cavitation retarded the formation of a passive layer on the electrode surface. Xu et al. [42] compared perfluorooctanoic acid removal using Ti/SnO₂-Sb/C-PbO₂ and Ti/IrO₂-RuO₂ anodes. Significant perfluorooctanoic acid removal was only observed when using the Ti/SnO₂-Sb/C-PbO₂ anode, and the application of ultrasound improved the degradation efficiency from 89% to nearly 99%. The authors measured the hydroxyl radical production using both anodes, finding

that the poor performance when using the Ti/IrO₂-RuO₂ anode was likely due to a lack of hydroxyl radical production. The significance of this radical on the degradation process was further highlighted when a HO[•] scavenger, *tert*-butyl alcohol, was introduced into the sonoelectrochemical experiments with Ti/SnO₂-Sb/C-PbO₂. At *tert*-butyl alcohol concentrations of 18 mM and 50 mM, the degradation efficiency was reduced from 98.7% to 43.9% and 25.2%, respectively. The proposed mineralisation mechanism was broadly in agreement with prior publications [43,186], with the first (and rate-limiting) step held to be direct oxidation of perfluorooctanoic acid at the anode. The authors postulated that the improvement offered by the combined sonoelectrochemical method was due to cavitation events causing higher temperatures and pressures near the electrode surface, thus enhancing the direct oxidation of perfluorooctanoic acid.

An interesting aside may be made at this point to consider why the

effectiveness of the electrooxidation of perfluorooctanoic acid was greatly enhanced by the application of ultrasound, whereas the study by Dionisio et al. [124] observed no improvement in the degradation of methyl paraben. The main difference between these studies is that the sonoelectrochemical degradation of methyl paraben was performed in a synthetic wastewater designed to simulate a real-world waste stream, thus containing several other species including sodium dodecyl sulfate, Na_2SO_4 , NaCl , H_2SO_4 , $\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$, $(\text{NH}_4)_2\text{SO}_4$, Na_3PO_4 and $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$. The authors pointed out that in a simple sulfate medium, it had already been shown that ultrasound enhanced the electrooxidation of methyl paraben, and sonication alone was enough to induce significant pollutant degradation [130]. In the synthetic wastewater, no sonochemical decomposition of methyl paraben could be obtained without coupling with electrolysis. This led them to conclude that, in the more complex synthetic wastewater, the ultrasound primarily influences the reaction through its mechanical effects, as opposed to the investigations into the breakdown of perfluorooctanoic acid, which highlighted the positive role of acoustically generated radicals. It is therefore pertinent to consider that the complexity of the medium may have a significant influence on the degradation of any given target pollutant (see also Section 3.5).

3.5. Sonoelectrochemical studies performed in real wastewater

As the previous example shows, mixed electrolyte systems present an added level of complexity to the functioning and optimisation of sonoelectrochemical systems for organic pollutant degradation. These challenges are in turn only magnified when decontamination of real-world waste streams is attempted [73,81]. For example, Yang et al. [40,187] employed a sonoelectrochemical method in conjunction with a Ru/Ir nano-coated anode, finding that these electrodes provided an increase of chemical oxygen demand removal by $\sim 24\%$ compared to electrodes without the coating (see also Section 2.1). It was proposed that sonication enhanced the pollutant degradation process through the cleaning and activation of the anode, improving the chemical catalytic activity of the nano-coated electrodes, and increasing hydroxyl radical diffusion through the bulk solution. Moreover, the wastewater treatment was accelerated under the action of an ultrasonic field due to high concentrations of chloride, from which highly oxidizing species could be generated (e.g. $\cdot\text{Cl}$, HClO , see Eqs (7)–(12)).

The beneficial impact of the presence of chloride was similarly highlighted by Tran et al. [153], who utilised a sonoelectrochemical method to treat wastewater containing the antibiotic chlortetracycline. Using a Ti-PbO_2 anode and a Ti cathode, a combined sonoelectrochemical method resulted in the degradation of up to 98% of the chlortetracycline in solution. A finding of significance was that the total organic carbon removal yield was low (37.2%), suggesting that a limited proportion of the organic material in the wastewater was completely mineralised in the process. The same researchers followed up this work by applying the same method to wastewater samples containing carbamazepine [152]. After performing a study to identify the optimal conditions (see Table 1), the authors recorded significant carbamazepine (93%), chemical oxygen demand (93%), total organic carbon (60%) and colour removal (86%). The toxicity of the effluent was also assessed using the bioluminescent bacterium *Aliivibrio fischeri*, with the results suggesting that the wastewater was completely de-toxified after sonoelectrochemical treatment.

Sonoelectrochemical oxidation has similarly been demonstrated as an effective treatment method for municipal wastewater. Llanos et al. [155] studied the disinfection of effluent from a municipal wastewater treatment plant in Spain using a boron doped diamond anode. Following the concentration of the microorganism *E. coli*, the authors first applied ultrasound alone (200 W). It was found that after an initial decrease in population, the level of *E. coli* plateaued; they deduced that, despite prior success in sono-disinfection being reported in the literature [188–190], the conditions in their study were not sufficient for full

disinfection of the wastewater effluent. However, when coupled with electrolysis, a synergistic effect was observed, which was ascribed to ultrasound preventing the agglomeration of the *E. coli*, making the population more susceptible to electrochemically-produced disinfectants such as hypochlorite and chloramines.

In a recent study, a hybrid electro-oxidation/coagulation treatment of sludge in municipal wastewater was examined [85]. Treatment and disposal of wastewater-activated sludge is expensive, and can account for up to 60% of the operational costs of a given facility [191–195]. The study used an electrode setup in which two anodes (one Ti-PbO_2 and the other a sacrificial Fe anode) were arranged in parallel with two stainless steel cathodes (Fig. 12); throughout the experiment the Fe anode was consumed, releasing positively-charged ions into solution [196]. These ions reacted with hydroxyl species generated through water reduction, to form a range of metal hydroxides and coagulants. Dissolved pollutants in the electrolyte then aggregated round these species, forming larger solid pieces which were easily separated and removed from the sample liquid as sludge. The authors evaluated the performance of the technique by considering factors such as the degree of degradation of the sludge, specific resistance to filtration, and the sludge moisture content. Applying sonication increased the degree of degradation achieved, although, crucially, the results indicated that the moisture content of the treated sludge was higher than when the process was performed in the absence of ultrasound. A consequence of this is that higher transportation and storage costs should be expected for sludge treated by this method over electro-oxidation/coagulation performed in the absence of ultrasound, which may limit its applicability.

More recently, Patidar and Srivastava [94] have applied this method

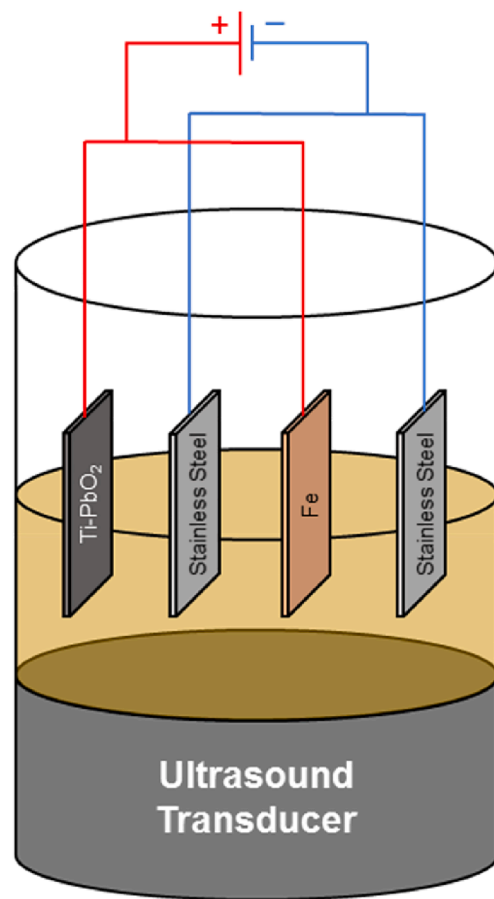


Fig. 12. A schematic of the reactor used in the treatment of municipal wastewater sludge via ultrasound assisted electro-oxidation/coagulation, adapted from ref. [85].

directly to samples collected from the compensation reservoir of a factory which produces various cosmetics such as soaps, lipsticks, and shampoo. Using a Ti/RuO₂ anode with Na₂SO₄ as a supporting electrolyte, a chemical oxygen demand removal efficiency of nearly 81% was achieved using a sonoelectrochemical approach under optimal conditions (Table 1). The researchers tested several other methods, finding that the chemical oxygen demand removal was higher for the sonoelectrochemical method than for sonication alone (6.8%), ultraviolet light (4.6%), purely electrochemical inputs (60.2%), and a combined electrochemical-ultraviolet method (74.9%). Furthermore, it was found that the energy consumption for the same degree of mineralisation using the sonoelectrochemical process was lower than for the electrochemical-ultraviolet approach (433.4 vs 540.5 kW per kg of chemical oxygen demand removed).

In terms of future prospects for sonoelectrochemical treatment of real-world waste streams, it should be noted that wastewater treatment systems normally integrate several different technologies in sequence [197]. The same strategy is also applicable to sonoelectrochemical processes, given the fact that a standalone sonoelectrochemical process is not necessarily capable of meeting all the treatment targets (i.e. removal of all pollutants and impurities). Although studies on the integration of sonoelectrochemical water treatment processes with other treatment technologies are lacking at the current time, reports on the integration of purely electrochemical processes into water treatment trains offer some idea of the pathway ahead [32,116,198]. For instance, electrochemical processes have been proposed as a pre-treatment step to improve the biodegradability of wastewater for subsequent biological processes and/or to help prevent membrane fouling [198]. Alternatively, electrochemical processes could be deployed as polishing (post-treatment) steps to degrade recalcitrant pollutants that fail to be removed by conventional technologies. Sonoelectrochemical treatments could well take on such roles, especially given the synergistic effects that are often observed between the electrochemical and sonochemical inputs, provided that the technical challenges associated with reactor design (especially incorporation of the ultrasound generation system) and the impact of ultrasound on the other technologies in the treatment train can be addressed.

4. Conclusions

Sonoelectrochemical oxidation for the degradation of pollutants is a growing field which is garnering ever increasing interest. In this review, we have introduced not only the core concepts behind this technique, but also the key factors behind its effectiveness, such as electrode materials, reactor design, choice of electrolyte, and operational parameters. Moreover, by presenting examples from the recent literature, we have demonstrated the potency of this technique for breaking down many different classes of compounds. Some key challenges must be addressed before this process can be fully adopted; perhaps the most significant is that these studies have to date all been performed in small batch reactors. The beneficial effects brought about by sonication are heavily influenced by reactor design (Section 2.2), and so it may not be possible to extrapolate the performance of all the studies discussed here to scaled-up systems. Another drawback to sonoelectrochemical approaches is their high energy consumption, and therefore cost: this may be abated to some extent by advancements in electrode technology [24,116,131] (and in some cases the application of ultrasound may lower the energy consumption of electrocatalytic pollutant conversion pathways through sono-electrocatalytic pathways [199]), but in any case such issues will need to be thoroughly investigated and addressed before widespread application of this technique can be achieved. These obstacles will no doubt be focal points for future study, and we hope that this review will go some way towards inspiring further strides in this field in the coming years.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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