



## Review Article

# Electrochemical technologies for the treatment of pesticides

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Nihal Oturan<sup>1</sup> and Mehmet A. Oturan<sup>1</sup>**Abstract**

Pesticides are used worldwide in large quantities to increase yield in agriculture. On the other hand, they are in general toxic/persistent organic pollutants presenting strong adverse effects to the environment and human health, including acute and chronic toxicity. Consequently, water polluted by pesticides should be treated efficiently before its release into receiving water bodies to protect the natural aquatic environment. Different methods have been used for the treatment of water contaminated by pesticides. Among them, electrochemical technology seems to be very efficient in removing pesticides from water. Therefore this review aims to provide an overview of the recent works on the treatment of pesticide wastewater using electrochemical technology with a special focus on electrochemical advanced oxidation processes that demonstrated high efficiency in the removal of various types of pesticides from contaminated water.

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Current Opinion in Electrochemistry 2021, 26:100677

This review comes from a themed issue on **Electrochemical Technologies for Wastewater Treatment**

Edited by **Carlos A Martínez-Huitle, Manuel Andres Rodrigo and Ignacio Sirés**

For complete overview about the section, refer [Electrochemical Technologies for Wastewater Treatment](#)

Available online 31 December 2020

<https://doi.org/10.1016/j.coelec.2020.100677>

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**Keywords**

Electro-Fenton, Pesticide, Electro-oxidation, Organic pollutants, Hydroxyl radicals, Wastewater treatment.

**Introduction**

The term pesticides cover a large variety of molecules used for improving agricultural productivity. With more than 4 million tons applied worldwide annually, they are among the most widespread xenobiotics released by anthropogenic activities [1]. It has been demonstrated that several pesticides present strong adverse effects on ecosystems (alteration of the biodiversity) and human health, including acute toxicity (accidental deaths by poisoning, particularly in developing countries) and chronic toxicity (even when exposed at low concentrations), as reported, in the case of India for example [2]. After being used on agricultural crops, pesticides reach the soil, air, and water bodies where their accumulation depends on their persistence and can reach significant concentrations in the different environmental compartments [1]. Regulatory authorities are trying to take into consideration this environmental issue by introducing standards for pesticide content in drinking water (e.g., in the European Union, 100 ng L<sup>-1</sup> for individual pesticides and 500 ng L<sup>-1</sup> for the sum of all pesticides detected [3]). Therefore, the removal of pesticides is currently an important challenge for environmental engineers working on soil remediation and water treatment, including drinking water, municipal wastewater, and some industrial wastewaters. The development of electrochemical processes might help to address this issue [4\*\*]. While the electrocoagulation process is able to separate some of the pesticides, electrochemical advanced oxidation processes (EAOPs) are widely recognized for their capacity to remove efficiently and in a nonselective way a very large range of pesticides from water [5]. EAOPs can also be applied for the removal of pesticides from groundwater and from the soil after a soil washing step [6\*\*]. The objective of this review is to provide a concise overview of the recent reports, including the advantages and

Table 1

## Compilation of recent works on the electrochemical treatment of pesticides in water.

Pesticide	Process	Cell configuration	Operating conditions	Removal efficiency	Energy consumption	Ref.
1,2-dichlorobenzene (0.1 mM = 14.7 mg L <sup>-1</sup> )	EF	Undivided cylindrical cell (230 mL), Nb/BDD anode (4 cm × 6 cm), carbon felt cathode (16 cm × 4 cm × 0.5 cm), with constant air supply	$i = 500$ mA, pH = 3, in Na <sub>2</sub> SO <sub>4</sub> 0.05 M, Fe <sup>2+</sup> = 0.1 mM	Complete degradation in 10 min $k^a = 294.6$ min <sup>-1</sup>	2.5 kWh (g-TOC) <sup>-1</sup>	[20]
Chloramben (herbicide) (1.19 mM = 245.1 mg L <sup>-1</sup> ) spiked in urban wastewater (TOC <sub>0</sub> = 15 mg L <sup>-1</sup> )	PEF <sup>c</sup>	Undivided cell (100 mL)	$j = 33.3$ mA cm <sup>-2</sup> , pH = 3.4, in Na <sub>2</sub> SO <sub>4</sub> 0.05 M, Fe <sup>2+</sup> = 0.05 mM, TOC <sub>0</sub> = 115 mg L <sup>-1</sup> , T = 35 °C	$k_{abs}^b = 1.61 \times 10^9$ M <sup>-1</sup> s <sup>-1</sup> >90% TOC removal (3 h) 96% degradation in 40 min	N/A	[21]
		Carbon-PTFE GDE <sup>d</sup> cathode (3 cm <sup>2</sup> ), BDD anode UVA lamp ( $\lambda = 365$ nm) Flow plant with a filter- press cell and a planar solar photoreactor, BDD anode (20 cm <sup>2</sup> ) and carbon-PTFE GDE cathode (20 cm <sup>2</sup> ), V = 2.5 L	$j = 50$ mA cm <sup>-2</sup> , pH = 3, in 0.05 Na <sub>2</sub> SO <sub>4</sub> , Fe <sup>2+</sup> = 0.5 mM, TOC <sub>0</sub> = 30 mg L <sup>-1</sup> , liquid flow rate = 200 L h <sup>-1</sup>	82% TOC removal (3 h) Time for complete degradation: 240 min for tebuthiuron and 120 min for ametryn.	2.1 kWh (g-TOC) <sup>-1</sup>	[16]
Tebuthiuron (0.18 mM = 41.1 mg L <sup>-1</sup> ) and ametryn (0.09 mM = 20.5 mg L <sup>-1</sup> ) from commercial formulations	SPEF <sup>e</sup>	Carbon-PTFE GDE <sup>d</sup> cathode (3 cm <sup>2</sup> ), BDD anode UVA lamp ( $\lambda = 365$ nm) Flow plant with a filter- press cell and a planar solar photoreactor, BDD anode (20 cm <sup>2</sup> ) and carbon-PTFE GDE cathode (20 cm <sup>2</sup> ), V = 2.5 L	$j = 50$ mA cm <sup>-2</sup> , pH = 3, in 0.05 Na <sub>2</sub> SO <sub>4</sub> , Fe <sup>2+</sup> = 0.5 mM, TOC <sub>0</sub> = 30 mg L <sup>-1</sup> , liquid flow rate = 200 L h <sup>-1</sup>	53% TOC removal (6 h) Complete degradation at 210 min	N/A	[29]
Imidacloprid (insecticide) (23.7 mg L <sup>-1</sup> ) in softened natural groundwater	AO-BDD	Undivided cylindrical cell (150 mL), Si/BDD anode, Fe cathode (both 10 cm <sup>2</sup> )	$j = 5$ mA cm <sup>-2</sup> , pH = 6.8, TOC <sub>0</sub> = 10 mg L <sup>-1</sup> , T = 25 °C, NO <sub>3</sub> <sup>-</sup> = 129.4 mg L <sup>-1</sup>	61.5% TOC removal (4 h)		
Lindane (10 mg L <sup>-1</sup> )	AO-BDD	Undivided cylindrical cell (230 mL), Nb/BDD anode (24 cm <sup>2</sup> ), carbon felt cathode (18 cm × 5 cm × 0.5 cm)	$j = 8.33$ mA cm <sup>-2</sup> , pH = 6.5, in Na <sub>2</sub> SO <sub>4</sub> 0.05 M, TOC <sub>0</sub> = 2.45 mg L <sup>-1</sup>	Total removal (10 min)	15 kWh (g-TOC) <sup>-1</sup>	[9 <sup>*</sup> ]
				$k^a_{tebuthiuron} = 0.098$ min <sup>-1</sup> , $k^a_{ametryn} = 0.26$ min <sup>-1</sup>		
				$k^a = 0.014$ min <sup>-1</sup>		
				$k^a = 0.25$ min <sup>-1</sup> 80% TOC removal (4 h)		

2,4-dichlorophenoxyacetic acid (20 mg L <sup>-1</sup> ) (herbicide)	PS <sup>f</sup> activation by AO-Blue TiO <sub>2</sub>	Undivided cell (100 mL), Blue TiO <sub>2</sub> anode, SS cathode	$j = 2.5 \text{ mA cm}^{-2}$ , pH = 3.5, PS = 30 mM	Complete degradation in 60 min $k^a = 0.07 \text{ min}^{-1}$ 83% TOC removal (2 h)	0.14 kWh m <sup>-3</sup>	[10]
Oxyfluorfen (100 mg L <sup>-1</sup> ) from commercial formulations (Fluoxil 24 EC)	EC <sup>g</sup>	Single-compartment flow cell (5 L-capacity), Fe anode, SS <sup>i</sup> cathode (both 100 cm <sup>2</sup> )	$j = 5 \text{ mA cm}^{-2}$ , pH = 7, in Na <sub>2</sub> SO <sub>4</sub> 0.02 M, TOC <sub>0</sub> = 72 mg L <sup>-1</sup> , $q = 0.6 \text{ L h}^{-1}$ , $Q = 0.836 \text{ A h dm}^{-3}$	99% depletion in 100 min	N/A	[18]
2,4-dichlorophenoxyacetic acid (10 mg L <sup>-1</sup> )	Solar PEC	3-electrode undivided cell (quartz), Ti/Blue TiO <sub>2</sub> photoanode (2.5 × 2.5 cm), SS cathode	E = 2.4 V vs SCE, pH = 5, in Na <sub>2</sub> SO <sub>4</sub> 0.05 M, sunlight (200 W Xenon lamp, 80 mW cm <sup>-2</sup> )	45% TOC removal (1.7 h) Total depletion in 120 min	N/A	[30]
2,4-dichlorophenoxyacetic acid (20 mg L <sup>-1</sup> )	H <sub>2</sub> O <sub>2</sub> -based electrochemical oxidation	3-electrode undivided cell (150 mL), Ti/RuO <sub>2</sub> -IrO <sub>2</sub> anode, N-doped graphene cathode (both 3 cm × 4 cm).	E = 4 V vs SCE, pH = 7, in 0.05 M Na <sub>2</sub> SO <sub>4</sub>	Total depletion in 60 min $k^a = 0.03 \text{ min}^{-1}$ $k^a = 0.04 \text{ min}^{-1}$		[26]
Monolinuron (herbicide) (0.1 mM = 21.5 mg L <sup>-1</sup> )	EF	Undivided cylindrical cell (230 mL), BDD anode (24 cm <sup>2</sup> ), carbon felt cathode, continuous air supply	$I = 500 \text{ mA}$ , pH = 3, in Na <sub>2</sub> SO <sub>4</sub> 0.05 M	88% TOC removal (8 h) Complete degradation in 10 min $k_{abs}^b = 3.1 \times 10^9 \text{ M}^{-1} \text{ s}^{-1}$	10 kWh (g-TOC) <sup>-1</sup>	[22]
Fenuron (0.1 mM = 16.42 mg L <sup>-1</sup> )	Photocatalytically-assisted electrochemical oxidation	Undivided cell (250 mL), dual anode: Ti/SnO <sub>2</sub> -Sb <sub>2</sub> O <sub>3</sub> /PbO <sub>2</sub> (electrocatalytic side), Ti/TiO <sub>2</sub> (photocatalytic side), SS cathode	$j = 30 \text{ mA cm}^{-2}$ , natural pH, in Na <sub>2</sub> SO <sub>4</sub> 0.05 M, $\lambda = 254 \text{ nm}$ .	98% TOC removal (8 h) 97.5% degradation in 60 min	N/A	[27]
1-chloro-2,4-dinitrobenzene (0.25 mM = 50.6 mg L <sup>-1</sup> )	AO-BDD	3-electrode undivided cell (150 mL), Si/BDD anode, graphite rod cathode, Hg/Hg <sub>2</sub> SO <sub>4</sub> reference electrode	$j = 30 \text{ mA cm}^{-2}$ , pH = 3, in Na <sub>2</sub> SO <sub>4</sub> 0.05 M, T = 30 °C	97.4% COD removal (4 h) 90% removal (230 min) $k^a = 0.01 \text{ min}^{-1}$	N/A	[31]
Glyphosate from commercial formulations (TOC = 100 mg L <sup>-1</sup> )	Cl <sup>-</sup> mediated AO	Undivided cell (100 mL), Ti/Ru <sub>0.36</sub> Ti <sub>0.64</sub> O <sub>2</sub> anode (14.76 cm <sup>2</sup> ), SS cathode (22.62 cm <sup>2</sup> )	$j = 40 \text{ mA cm}^{-2}$ , pH = 3, NaCl = 0.15 M	62% TOC removal (3.8 h) 60% TOC removal (3 h)	1 kWh (g-TOC) <sup>-1</sup> (for solutions with the pure chemical)	[32]

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Table 2

## Examples of application of electrochemical technology to the treatment of pesticides in soils and groundwater.

1- Target pollutant 2- Washing solution	Process	Cell configuration Operating conditions	Removal efficiency	Ref.
1 Atrazine (spiked clay)	AO	Continuous circulation (200 L h <sup>-1</sup> ) of the effluent (1 L) in a flow-by cell using BDD anode and steel (diameter = 10 cm). $I = 30 \text{ mA cm}^{-2}$	Complete degradation of atrazine and sodium dodecyl sulfate and >90% TOC removal at 30 Ah L <sup>-1</sup> .	[36]
2 Sodium dodecyl sulfate (100–5000 mg L <sup>-1</sup> ) and NaHCO <sub>3</sub> (500 mg L <sup>-1</sup> )				
1 Oxyfluorfen (spiked clay)	AO and sono-A	Continuous circulation (200 L h <sup>-1</sup> ) of the effluent (1 L) in a flow-by cell using BDD anode and steel cathode (diameter = 10 cm). $I = 30 \text{ mA cm}^{-2}$ Auxiliary tank for irradiated experiments (24 kHz, 250 W L <sup>-1</sup> )	Complete degradation of oxyfluorfen at 25 Ah L <sup>-1</sup> for AO and 40 Ah L <sup>-1</sup> for sono-AO. >90% TOC removal at 30 Ah L <sup>-1</sup> .	[42]
2 Sodium dodecyl sulfate (100–5000 mg L <sup>-1</sup> ) and NaHCO <sub>3</sub> (500 mg L <sup>-1</sup> )				
1 Lindane (spiked clay soil)	AO	Continuous circulation (60 L h <sup>-1</sup> ) of the effluent (1 L) in a flow-by cell using BDD anode and steel cathode (diameter = 10 cm). $I = 50 \text{ mA cm}^{-2}$	Complete degradation of lindane at 15 Ah L <sup>-1</sup> or 80 Ah L <sup>-1</sup> depending on extracting agent concentration.	[43]
2 Sodium dodecyl sulfate (100–5000 mg L <sup>-1</sup> ) and NaHCO <sub>3</sub> (500 mg L <sup>-1</sup> )				
1 Clopyralid	EF	Batch undivided cylindrical glass reactor (800 mL) with Pt anode (32 cm <sup>2</sup> ) and carbon felt cathode (112 cm <sup>2</sup> ). Continuous air bubbling. [Fe <sup>2+</sup> ] <sub>0</sub> = 0.1–5 mM pH = 3 $I = 50\text{--}300 \text{ mA}$	80% degradation of clopyralid, 30% mineralization and biodegradability increase after 480 min of treatment at 200 mA.	[44]
2 Synthetic groundwater				
1 Phenanthrene (not a pesticide)	EF and AO	Batch undivided cylindrical glass reactor (400 mL) with Pt, DSA or BDD anode and carbon felt cathode (150 cm <sup>2</sup> ). [Na <sub>2</sub> SO <sub>4</sub> ] = 0.15 M [Fe <sup>2+</sup> ] = 0.2 mM and air bubbling for EF. $I = 500\text{--}2000 \text{ mA}$	Complete removal of degradation after 240 min at 500 mA for EF-BDD ( $k_{\text{app}} = 0.012 \text{ min}^{-1}$ ) and 9% TOC removal.	[38]
2 Cyclodextrin (9000 mg L <sup>-1</sup> )				
1 Phenanthrene or several PAHs (not pesticides)	AO	Batch undivided cylindrical glass reactor (400 mL) with BDD anode and stainless steel	Selective degradation of target pollutants. 75–90% removal of PAHs after 23 h at 2.1 mA cm <sup>-2</sup> , while	[40**]

(continued on next page)

Table 2 (continued)

1- Target pollutant 2- Washing solution	Process	Cell configuration Operating conditions	Removal efficiency	Ref.
2 Tween® (6550 mg L <sup>-1</sup> )	80	cathode (150 cm <sup>2</sup> ). [Na <sub>2</sub> SO <sub>4</sub> ] = 0.05 M.  I = 2.1–42 mA cm <sup>-2</sup>	the extraction capacity was only 5% lower than the fresh one.	

The use of accessible carbonaceous cathode materials (i.e., carbon felt, carbon cloth, carbon fibers, etc.) constitutes a technical advantage of EF and makes the process economically attractive. In contrast, BDD is still a costly material restraining the application of AO at a large-scale [24,25\*].

Different pesticides have been used as model pollutants to evaluate the performance of new electrode materials. For example, an N-doped graphene cathode for H<sub>2</sub>O<sub>2</sub> production and its activation was successfully tested for the degradation of 2,4-dichlorophenoxyacetic acid [26], while a dual anode consisting of one layer of SnO<sub>2</sub>–Sb<sub>2</sub>O<sub>3</sub>/PbO<sub>2</sub> and one of TiO<sub>2</sub> deposited on the opposite sides of a Ti plate was used to treat the herbicide fenuron by photocatalytically assisted AO [27]. The development of high-surface-area and durable electrode materials is an active research field with great improvement opportunities.

On the other hand, most of the investigations have been conducted using aqueous solutions of the target compounds (some of them using commercial formulations) to obtain important information about the kinetics, mechanisms, and optimal experimental conditions for the degradation/mineralization of such pollutants. However, the effect of the matrix in real wastewaters is not considered in such works. Hence, more investigations using real water sources contaminated by pesticides are highly recommended in order to evaluate the influence of the matrix (additives, other organics, as well as inorganic compounds) on the process performance [25\*]. In this sense, the presence of radical scavengers may significantly affect the degradation/mineralization efficiency. Additionally, the concentrations of target pollutants in synthetic solutions are generally orders of magnitude higher than the concentrations in real water sources [28], which may affect the efficiencies under real scenarios, especially considering the mass transport limitations to the electrode surface inherent to electrochemical processes.

Another important point to keep in mind is that most studies on the electrochemical treatment of pesticides have been carried out in small laboratory-scale

devices with capacities rarely exceeding 1 L. With this respect, the design of electrochemical reactors with the capacity to treat greater volumes is fundamental for the application of such treatment methods at an industrial scale. Large-scale systems would give a more reliable assessment of both investment and operating costs of electrochemical processes (operating costs tightly related to electrical energy consumption).

### Applications for the treatment of pesticides in soil

Diffuse pollution of soils is a very complex environmental issue because of the huge amount of soils that might be concerned for remediation and the great complexity of treating contaminated soils. Electrochemical technologies cannot be considered as sustainable solutions in such a case. However, manufacturing and handling of pesticides can lead to accidental release and create localized acute contamination, for which the application of electrochemical technologies might be suitable.

First, electrokinetic processes based on the application of an electric field in the soil present the great interest to avoid the use of any chemical compounds and to be applied *in situ*. It has been reported at the lab-scale that it is possible to recover pollutants in catholyte and anolyte wells by dragging with the electroosmotic flux or by electromigration of ionic compounds [19]. However, it has been highlighted that the influence of these mechanisms is very low when applying this process at a pilot scale [19]. As high current intensities have to be applied, the controlling mechanism becomes the electric heating of the soil, which influences pollutant volatilization from the soil. Current studies focus mainly on the combination with other processes (e.g., phytoremediation or the use of additives such as oxidants) [33\*,34] or by efficiently implementing this technology at pilot-scale (optimization of powering control, inversion of electrode polarity, etc.) [33\*,34,35].

Second, a treatment strategy involving an electrochemical process as a post-treatment step can be applied for treating soil washing solutions [6\*\*]. Soil washing (as the first step) aims at transferring pesticides

from the soil-sorbed fraction to the aqueous phase. Extracting agents (cyclodextrins or surfactants such as Tween 80, Triton X 100, or sodium dodecyl sulfate) can be used for improving the transfer of hydrophobic pesticides. The soil washing solution is, therefore, a complex mixture of all compounds that can be mobilized, including target pesticides, soil organic matter, inorganic species, fine particles, and extracting agents [6\*\*]. Several studies have reported that EAOPs are able to remove the organic load of these effluents [36,37\*]. Recent studies are reported in Table 2. The possibility of combination with a biological treatment has also been assessed for improving the cost-effectiveness [38,39], and few studies focused on selective electro-oxidation of target pollutants in order to be able to reuse extracting agents [39\*\*,40\*,41\*]. However, further studies are required in order to scale up such treatment for field-scale applications and for assessing the sustainability of this approach.

### Challenges for full-scale applications

There are several challenges for upscaling the electrochemical technologies regarding the removal of pesticides, especially about the reactor design.

When the option consists of treating pesticides from urban wastewater or natural water, their concentrations in solution is very low (from  $\text{ng L}^{-1}$  to  $\mu\text{g L}^{-1}$ ) [4\*]. It means that it is required to strongly favor the contact between the pollutant and the electrodes in AO and/or to enhance the promotion of the homogeneous oxidation with the EF process [45]. This will enhance the faradaic yield while keeping high pesticide removal yields. Moreover, these effluents generally have a low electric conductivity [4\*], which means that the reactor design needs to be also adapted in order to avoid adding a supporting electrolyte. Microfluidic thin-film electrochemical reactors have emerged as a possible response towards these issues by intensifying the transport of species and reducing the ohmic resistance [46–48]). Further studies are needed at a large scale in order to validate the lab-scale systems.

When the pesticides are solubilized in soil washing/flushing effluent, their concentrations in a solution can be increased to  $\text{mg L}^{-1}$  range [28]. Still, the faradaic yield remains low because the extracting agent represents the main organic source. For making it economically viable at a larger scale, the recovery of the extracting agent has to be considered, while the selective removal of the contaminants should be effective. Combined treatments in hybrid or sequenced reactor(s) required supplementary proofs at high technology readiness levels.

In electrokinetics systems, lab-scale experiments could not predict well the pilot and large-scale applications

[33\*]. The difficulties remain in the ability to have geometrical and operational similarity between the setups [33\*], which makes changing the distribution of parameters (pH, conductivity, temperature, pollutants, and ions concentrations) and the mechanisms involved [19,49]. Furthermore, the soil characteristics (electric conductivity, granulometry, age of pollution, and so on) vary a lot from one sample to another, which makes difficult the extrapolation of the results for upscaling studies. Additional large-scale studies need to be performed in a comparable way by implementing dimensional analysis.

### Conclusions and future perspectives

Electrochemical processes stand out as promising technologies for the treatment of harmful organic pollutants (pesticides, synthetic dyes, pharmaceutical residues, and so on) in contaminated water and soils. Particularly, EAOPs present great advantages due to their capacity to remove, in a nonselective way, a large range of pesticides, which can be degraded and even fully mineralized in order to avoid the presence of toxic by-products remaining in the treated solution. Besides application to contaminated water, they can also be employed for the removal of pesticides in soil washing effluents. The next challenge for further development of electrochemical technology and particularly of EAOPs is clearly related to full-scale applications, including the design of suitable reactors and the application to real effluents. This step is required in order to conclude on the sustainability and cost-effectiveness of these processes.

### 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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