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Advanced hybrid electro-separation/electro-conversion systems for wastewater treatment, reuse and recovery: compromise between symmetric and asymmetric constraints

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Abstract

Wastewater treatment with reuse and recovery of value-added compounds for valorization is of rising interest, and the combination of electro-separation and electro-conversion processes could be a promising solution to both environmental and resource availability problems. The more recent concomitant development of both electrochemical advanced oxidation processes and materials with new properties make the older electro-separation technologies regain visibility and interest. The electrofiltration/electrooxidation or electroreduction, electrosorption/electrooxidation, electrocoagulation/electro-Fenton, electroprecipitation/electrooxidation and electrodeposition/electrooxidation have been particularly critically reviewed. The conventional flow-by or flow-through parallel-plate and concentric cylinders design do not suffice to face the antagonist requirements in such simultaneous multiple electroproccesses. Innovative designs are needed and emerging concepts such as reactive electro-mixing are a possibility. Further modeling and scale-up studies based on revised theory are required in the future.

Keywords:

Anodic oxidation; peroxi-coagulation; electro-sorption; reactive electrochemical membrane; reactive electro-mixing; reactor design

Highlights

- Electro-separation/electro-conversion combination in simultaneous operation
- Competition between symmetric and asymmetric constraints
- Need for innovative design that consider antagonist requirements

1. Introduction

The rapid increase in water stress due to climate change while water demand is rising represents a major concern to the environment and to social structures [1]. The United Nations has identified water shortages as one of the biggest challenges to overcome in the next few decades [2]. One of the solutions is to adopt a water reuse strategy [3], which may also include the valorization of value-added inorganic and organic compounds often contained in municipal and industrial wastewaters [1]. In this context, advanced physico-chemical treatments have been developed as complementary or alternative techniques to conventional biotechnologies [4–7]. Among them, electrochemical systems could take advantage of their modularity, flexibility and sustainability [8,9].

Hybrid reactors that combine electro-separation systems with electro-conversion technology are attracting attention in wastewater treatment because of possible synergies in their application with reductions in footprint [10–13]. Electro-separation systems can separate selectively or not the compounds from the solution in order to recover value-added components and/or to avoid the release of electrogenerated by-products [14,15]. In addition, the electro-conversion permits elimination of worthless pollution and/or its conversion into valuable molecules [9,16,17]. Implementing this combination in a hybrid system can also concentrate the pollutants by electro-separation, while porous electrodes can be regenerated by electro-conversion [18,19]. However, these more complex systems require adequate design for an optimized process. Some of the developed technology lacks electrochemical engineering considerations. Therefore, the goal of this review paper is to list constraints in, and assemble tools for the design of scalable multifunctional electrochemical systems for applications in wastewater treatment, reuse and recovery.

2. Emerging hybrid electro-separation/electro-conversion systems for wastewater treatment, reuse and recovery

The hybrid electro-separation/electro-conversion combinations are critically discussed in this section and the main recent papers are described in Table 1 [10–13,18–29], while examples of reactor setup for each combination are presented in Fig. 1 [10,11,13,23–25].

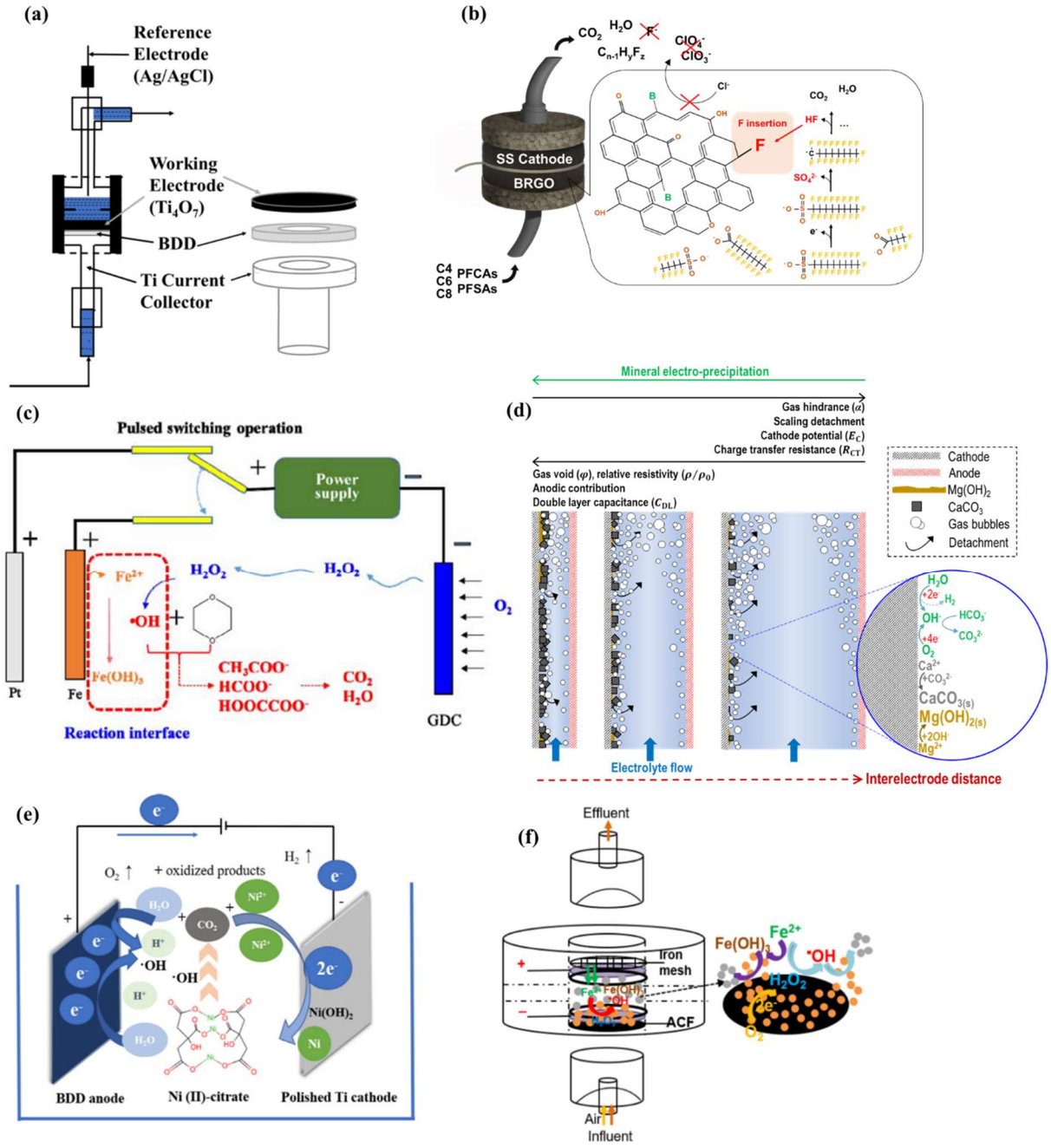

Fig. 1. Hybrid reactor setup implementing electro-separation/electro-conversion combinations in simultaneous operation: (a) electrofiltration/electrooxidation in flow-through parallel-plate system (Reproduced with permission from [10]. Copyright 2020, American Chemical Society), (b) electrosorption/electrooxidation in flow-through parallel-plate system (Reproduced with permission from [11]. Copyright 2022, Elsevier), (c) electrocoagulation/electro-Fenton in stirred tank reactor with electrodes in parallel (Adapted with permission from [13]. Copyright 2021, IWA Publishing), (d) electroprecipitation/electrooxidation in flow-by parallel-plate system (Adapted with permission from [23]. Copyright 2022, Elsevier), (e) electrodeposition/electrooxidation in stirred tank reactor with electrodes in parallel (Reproduced with permission from [24]. Copyright 2022, Elsevier), (f) Electrosorption/electrocoagulation/electro-Fenton in flow-through parallel-plate system (Reproduced with permission from [25]. Copyright 2021, Elsevier).

2.1. Electrofiltration/electrooxidation or electrofiltration/electroreduction

Electrified membranes, also called reactive electrochemical membranes (REMs), have been developed to combine electrofiltration with electrooxidation or electroreduction in order to enhance mass transport of organic and inorganic pollutants and concentrate them in the permeable electrode material, where they can be degraded and mineralized (Table 1) [30–32]. The porous anode material is usually coated with elements such as boron (boron-doped diamond (BDD)) (Fig. 1a) [10], tin oxide (SnO_2-Sb) [27], β-PbO_2 or sub-stoichiometric titanium oxide (Ti_4O_7) [10,26] that enable an increase its O_2 evolution overvoltage. This property allows electrogeneration of physisorbed hydroxyl radicals ($^{\bullet}OH$) from H_2O oxidation in the vicinity of the anode, which occurs within nanometers from the surface due to the very short lifetime (10^{-9} s) of $^{\bullet}OH$ [33,34]. These radicals are very powerful oxidants that can degrade quasi non-selectively the organic pollutants in wastewater [35]. The electrofiltration/electrooxidation combination allows degradation of pharmaceutical pollutants such as ciprofloxacin at very high level (92%) and in a single pass [27], while longer residence times (5-fold) are required without the combination. In the meantime, electroreduction of oxyanions (e.g., ClO_3^-, ClO_4^-, BrO_3^-, NO_3^-) becomes an important consideration, as they are known to occur during advanced electrooxidation by these radicals in wastewater containing halogenated and/or nitrogenous ions [36,37]. In this context, REM has also been tested and combined with electroreduction by implementing oxorhenium complex coated on Ti_4O_7 cathodes for the elimination of perchlorate from wastewater [28]. Perchlorate could be reduced by 21% with 99% selectivity towards Cl^-, due to the electrofiltration/electroreduction combination.

REM systems present the advantage of decreasing the fouling compared to filtration alone, to a certain extent [38]. Nevertheless, the average pore diameters of such anodic filters are usually in the micrometer range [26,27], meaning that currently REM is only be suitable for micro- or ultra-filtration, but not for nanofiltration or reverse osmosis [30]. Moreover, the mechanism responsible for compound retention within the material would be more electrosorption than electrofiltration under these conditions [30]. This means that additional key parameters presented in sub-section 2.2 would also need to be considered for this hybrid system optimization.

2.2. Electrosorption/electrooxidation

Electrosorption describes the enhancement of adsorption by polarizing conductive materials at low electrode potentials, usually between 0.4 and 1.4 V/standard hydrogen electrode (SHE), in order to remain in the capacitive current domain [14,15,39]. Under such conditions, no faradaic reaction should occur (e.g. H_2O oxidation and reduction), while the double-layer capacitance is maximized [14,39]. This latter parameter along with mass transport phenomena are the major factors affecting the electrosorption efficiency [14]. This technology arose from interest in removing inorganic salts in brine water for its desalination at low energy demand compared to technology currently applied (e.g., reverse osmosis) [15]. This technique has also been applied to remove selectively and recover valuable organic compounds such as phenolic compounds [19] or benzoate as representative organic anions [40].

Electrosorption has been combined with electrooxidation to eliminate inorganic (arsenic (AS(III) [29]) and organic pollutants (e.g., naproxene [20], per- and polyfluoroalkyl substances (PFASs) (Fig. 1b) [11], N,N-diethyl-meta-toluamide (DEET), phenolic compounds [19], iopromide, carbamazepine and diatrizoate [18]) from wastewater (Table 1). Synergies have been obtained with this hybrid combination. For example As(III) was electrosorbed and accumulated (100 mg-As g(adsorbent)$^{-1}$) in the electrolyzer, and subsequently oxidized into a less hazardous form of As (As(V)) (>90% efficiency) [29]. In a similar manner, electrosorption permits concentration of persistent organic pollutants having a low initial concentration in water (0.2 µmol L^{-1} each), which can then be degraded by electrooxidation with synergy of removal ranging from 30 to 57% [11,18]. Interestingly, the amount of chlorinated by-product released in solution was diminished with the hybrid system, due to the electrosorption of such undesirable compounds.

However, electrosorption and electrooxidation do not operate over the same optimal potential range, which needs further innovative reactor design as discussed in section 3.

2.3. Electrocoagulation/electro-Fenton

Electrocoagulation corresponds to the electrogeneration of metal ions in the bulk electrolyte by anodic dissolution of the polarized metal, which is then hydrolyzed in water [41]. This permit the release of various coagulants in the electrolyte such as hydroxide sludges, depending on the

solution pH. The pollutants in wastewater - suspended solids to a large extent - can then be removed mainly by precipitation, coprecipitation and adsorption on precipitates [42].

Iron is one of the sacrificial anodes mostly used in electrocoagulation and has been combined with the electrogeneration of H_2O_2 from O_2 reduction at the cathode to form $^{\bullet}OH$ through the Fenton reaction, namely the peroxicoagulation process [12,43]. This combination allows simultaneous electrocoagulation and electro-Fenton within the same reactor with synergies in terms of energy consumption and removal of organic pollutants (Table 1) [12,13,21]. For instance, percentage removal higher than 92% could be reached for acrylonitrile [12], 1,4-dioxane (Fig. 1c) [13] and 2,4-dichlorophenoxiacetic acid [21] with the combination.

The hybrid combination of electrocoagulation or peroxicoagulation with advanced electrooxidation could be interesting in simultaneous operation to enhance the process efficiency, but these processes require a different anode material. This is the reason why the existing electrocoagulation/electrooxidation combinations are operated in a sequential mode instead of simultaneously [44,45]. Thus, new designs are required for such more complex hybrid systems as discussed in section 3.

2.4. Electroprecipitation/electrooxidation

Electroprecipitation phenomena occur principally on cathode surfaces due to local alkalization from O_2 and H_2O reduction reactions [23,46,47]. This local drastic increase in HO^- ions ensures ions like magnesium (Mg^{2+}), calcium (Ca^{2+}) and carbonates (CO_3^{2-}) precipitate mainly as hydroxides (e.g., $Mg(OH)_2$) and calcium carbonate ($CaCO_3$) [48–51]. This technology has been applied widely for water softening processes. More recently, the interest in this system for calcium phosphate precipitation and subsequent phosphorus recovery has been assessed [52–54]. The possible synergy that could be obtained between electroprecipitation and advanced electrooxidation using a BDD anode has been further studied (Table 1) [22,23]. It was highlighted that an anodic contribution on electroprecipitation phenomena could appear when interelectrode distances are varied from 50 to 3000 µm (Fig. 1d) [23]. Still, further studies are necessary at different current densities and in the presence of organic pollutants to carry out the whole combination efficiently.

2.5. Electrodeposition/electrooxidation

In contrast to electroprecipitation, electrodeposition represents direct electron transfer between the electrode and the metal element to be deposited on the polarized material [55–57]. This technique is used widely in extraction and metal plating for subsequent metal recovery, with high purity attained in many cases [55–57]. Simultaneous electrodeposition and electrooxidation has been investigated recently (Table 1). Ni(II)-citrate was removed successfully (100% removal) by electrooxidation at the BDD anode, while metallic nickel was recovered on a titanium cathode by electrodeposition with 95% of purity (Fig. 1e) [24]. More studies are needed to assess the selectivity and efficiency of this approach with real effluents.

2.6. Hybrid multiple simultaneous electroprocesses

The combination of multiple electroprocesses higher than two and operated simultaneously in a single reactor has been barely examined as yet. Electrosorption has been combined with peroxicoagulation recently in a flow-through cell design (Table 1 and Fig. 1f) [25]. The coupling improved by 10-fold the electrosorption capacity compared with that of electrosorption alone, while 93% of dye removal was reached. However, electrosorption in this system was associated more with a pseudo-capacitive electrosorption, because it was performed under faradaic conditions. This means that the double-layer capacitance was not optimized. Thus, the conventional design of electrochemical reactors limits the possibility to operate with multiple simultaneous electroprocesses, knowing the constraints that need to be addressed as discussed in section 3.

1 **Table 1.** Main recent hybrid combination of electroseparation with electroconversion processes for wastewater treatment.

Type of electroseparation/ electroconversion system and design	Targeted compound(s)	Characteristics of electrolyte	Electrode materials and distance	Studied parameters	Efficiency	Reference
Electrofiltration/ Electrooxidation in a hybrid undivided cylindrical parallel-plate flow-through cell, in simultaneous operation	Resorcinol	Synthetic wastewater containing 0.006-0.01 mol L^{-1} of KH_2PO_4, 0-0.05 mol L^{-1} NaCl and 0.001 mol L^{-1} of resorcinol	Anode: Ti_4O_7 or BDD Cathode: BDD	Initial electrolyte concentration, electrolysis time, anode material	68% of resorcinol mineralization	[10]
Electrofiltration/ Electrooxidation in a hybrid undivided tubular flow-through cell, in simultaneous operation	Carbamazepine	Real secondary effluent from urban WWTP spiked with carbamazepine at 100 µg L^{-1}	Anode: Ti_4O_7 Cathode: carbon felt	Current density, flux, electrolysis time	>98% degradation of carbamazepine and 70% of mineralization with an energy consumption of 0.15 kWh g^{-1} of TOC removed	[26]
Electrofiltration/ Electrooxidation in a hybrid undivided cylindrical parallel-plate flow-through cell, in simultaneous operation	Ciprofloxacin	Synthetic wastewater containing 0.1 mol L^{-1} of Na_2SO_4 and 10 µg L^{-1} of ciprofloxacin	Anode: porous Sb-SnO_2 Cathode: Ti mesh	Reactor configuration (flow-by or flow-through), electrolysis time, initial concentration of pollutant, flux,	92% of ciprofloxacin removal in single pass (0.33 kWh m^{-3})	[27]
Electrofiltration/ Electroreduction in a hybrid undivided cylindrical parallel-plate flow-through cell, in simultaneous operation	Perchlorate	Synthetic wastewater containing 0.01 mol L^{-1} of KH_2PO_4 and 0.005 mol L^{-1} of $NaClO_4$	Anode: BDD Cathode: oxorhenium complex coated on Ti_4O_7	Electrode potential, flux, solution pH	21% of perchlorate reduction with 99% Cl^- selectivity	[28]

Electrosorption/ Electrooxidation in a hybrid undivided flow-by cell, with simultaneous operation	Arsenic (As(III))	Real secondary effluent from urban WWTP spiked with As(III) at 100 µg L^{-1}	Working electrode: poly(vinyl)ferrocene (PVF)/carbon nanotube composite Counter electrode: poly-TEMPO-methacrylate (PTMA) Electrode potential: 0.8 V/Ag-AgCl for 2 h	Electrolysis time, working electrode material, number of cycles of regeneration	37% of As(III) electrosorbed 93% of As(III) conversion into As(V) 0.45 kWh mol-As^{-1} (electrosorption + electroconversion)	[29]
Electrosorption/ Electrooxidation in a hybrid undivided stirred tank design with three-electrode system, in sequenced operation	Naproxene	10 mg L^{-1} naproxene and 0.05 mol L^{-1} of KNO_3 solution at pH 7	Working electrode: packed activated carbon with stainless steel as current collector Counter electrode: stainless steel mesh Electrode potential: 1.2-1.4 V/NHE Interelectrode distance: 2 cm	Electrode potential, electrosorptin time, regeneration (reactivation) time	Regeneration (reactivation) increased electrosorption capacity (83%) at 0.7 V/NHE	[20]
Electrosorption/ Electrooxidation in a hybrid undivided cylindrical flow-through cell, in simultaneous operation	Per- and polyfluoroalkyl substances (PFASs)	PFASs (0.2 µmol L^{-1} each) in 10 mmol L^{-1} phosphate buffer (pH around 7.1)	Anode: graphene sponge with stainless steel as current collector Cathode: stainless steel sponge	Flow rate, anode potential, electrolysis time	67% of removal (35% by electrosorption; 32% by electrooxidation) at 230 A m^{-2}, leading to 10.1 kWh m^{-3}	[11]
Electrosorption/ Electrooxidation in a hybrid undivided flow-through cell with packed-bed material, in sequenced operation	N,N-diethyl-meta-toluamide (DEET), iopromide, carbamazepine and diatrizoate	Real secondary effluent from urban WWTP spiked with DEET, iopromide, carbamazepine and	Anode: mesh Nb/ BDD Cathode: stainless steel	Current density	DEET presented the highest synergy, ranging from 40% to 57%, followed by iopromide (22–46%),	[18]

		diatrizoate (0.2 µmol L^{-1} each)	Granular activated carbon between cathode and anode		carbamazepine (15–34%) and diatrizoate (4–30%)	
Electrosorption/ Electrooxidation in a hybrid cylindrical flow-through cell with packed-bed electrode, in simultaneous operation	Phenolic compounds	Real olive mill wastewater effluent with total phenolic compounds at 10.6 g L^{-1}	Anode: graphite rod Cathode: alginate/powdered activated carbon composite with a graphite rod as current collector	Electrolysis time, activated carbon load, electrode potential, effluent composition	307 mg g^{-1} with electrosorption and electrooxidation occurred simultaneously at a cathode potential below -1.2 V/Ag-AgCl	[19]
Electrocoagulation/ Electro-Fenton (= peroxicoagulation) in a hybrid undivided stirred tank reactor with electrodes in parallel, in simultaneous operation	Acrylonitrile	Synthetic wastewater containing 0.05 mol L^{-1} of Na_2SO_4 and 50-500 mg L^{-1} of acrylonitrile	Anode: iron plate Cathode: graphite felt coated with carbon black Interelectrode distance: 2 cm	Electrolysis time, current density, pH of solution, initial concentration of pollutant	More efficiency with the combination (92% of acrylonitrile removal) Less energy with combination (3.08 kWh kg-acrylonitrile^{-1})	[12]
Electrocoagulation/ Electro-Fenton (= peroxicoagulation) in a hybrid undivided stirred tank reactor with electrodes in parallel, in simultaneous operation	1,4-dioxane	Synthetic wastewater containing 0.1 mol L^{-1} of Na_2SO_4 and 200 mg L^{-1} of 1,4 dioxane at pH 5	Anode: iron mesh Cathode: gas diffusion electrode with carbon black layer and a supporting layer of stainless steel mesh Interelectrode distances: 2-3.5 cm	Current density, pulsed switching frequency, electrolysis time	More efficiency with the combination (95% of pollutant removal) Less energy with combination (7.8 kWh kg-pollutant^{-1})	[13]
Electrocoagulation/ Electro-Fenton (= peroxicoagulation) in a hybrid undivided stirred tank reactor with electrodes in parallel, in simultaneous operation	2,4-dichlorophenoxiacetic acid	Synthetic wastewater containing 0.1 mol L^{-1} of Na_2SO_4 and 50-300 mg L^{-1} of 2,4-dichlorophenoxiacetic acid	Anode: iron plate Cathode: graphite felt coated with carbon black or natural air diffusion electrode	Electrode material, electrolysis time, current density, pH of solution,	More efficiency with the combination (100% of pollutant removal: 59.4% with	[21]

			Interelectrode distance: 1 cm	initial concentration of pollutant	electrocoagulation and 40.6% with electro-Fenton) Less energy with combination (12 kWh kg-pollutant^{-1})	
Electroprecipitation/ Electrooxidation in a hybrid undivided parallel-plate flow-by cell, in simultaneous operation	Magnesium, calcium, carbonates	Synthetic wastewater containing 0.02-0.10 mol L^{-1} of Na_2SO_4 and 150 mg L^{-1} of Ca^{2+}, 5 mg L^{-1} of Mg^{2+}, 60 mg-C L^{-1} of HCO_3^-/CO_3^{2-}	Anode: BDD Cathode: stainless steel Interelectrode distance: 500 µm	Electrolysis time, current density, effluent composition	Electrolyte composition influence co-electroprecipitation, while anodic oxidation did not influence it in applied condition	[22]
Electroprecipitation/ Electrooxidation in a hybrid undivided parallel-plate flow-by cell, in simultaneous operation	Magnesium, calcium, carbonates	Synthetic wastewater containing 0.02-0.10 mol L^{-1} of Na_2SO_4 and 150 mg L^{-1} of Ca^{2+}, 5 mg L^{-1} of Mg^{2+}, 60 mg-C L^{-1} of HCO_3^-/CO_3^{2-}	Anode: BDD Cathode: stainless steel Interelectrode distances: 50-3000 µm	Electrolysis time, interelectrode distance, cathode potential	Maximal electroprecipitation at 100 µm gap, with possible anodic oxidation contribution that increased cathodic electroprecipitation	[23]
Electrodeposition/ Electrooxidation in a hybrid undivided stirred tank reactor with electrodes in parallel, in simultaneous operation	Ni(II)-citrate	Synthetic wastewater containing 0.08 mol L^{-1} of electrolyte and 75 mg L^{-1} of Ni(II)-citrate	Anode: BDD Cathode: polished titanium	Initial nickel citrate concentration, current density, initial pH, interelectrode distance, electrolyte type, initial electrolyte concentration	100% of Ni(II)-citrate removal with 72% of metallic nickel recovery (95% of purity)	[24]

Electrosorption/ Electrocoagulation/ Electro-Fenton in a hybrid undivided parallel-plate flow-through cell, in simultaneous operation	Orange II, 2,4-dichlorophenoxyacetic acid, phenol and methylene blue	Synthetic wastewater containing 0.05 mol L^{-1} of Na_2SO_4	Anode: Iron sheet Cathode: activated carbon fiber Interelectrode distances: 8 mm	Solution pH, flow rate, initial concentration of organics, applied voltage	93% of dye removal and 1043 mg g^{-1} of electrosorption capacity (10 times higher than without combination)	[25]

2 Abbreviations: BDD, boron-doped diamond; NHE, normal hydrogen electrode; WWTP, wastewater treatment plant

3. Constraints with simultaneous multiple electroprocess implementation and emerging design considerations

3.1. Symmetric versus asymmetric constraints

In electrochemical systems, symmetric constraints need to be considered for optimal efficiency, as mentioned in renowned textbooks [58–62] and review papers [63,64] in the field of electrochemical engineering. The current and potential distributions are important criteria that need to be kept as homogenized as possible [65,66]. This is to ensure the same efficiency of electrosorption or electro-conversion on the entire active surface of the electrode so that it is possible to make full use of the electrode surface. In practice, symmetric geometries, such as parallel-plate cells or concentric cylinders with non-porous plane materials are usually favored [58,63]. However, porous materials are required in some electro-separation processes as mentioned in section 2, while other electroprocesses benefit from porous electrodes to enhance mass transfer by implementing a flow-through configuration. These configurations could result in heterogeneous distributions of current and potential within the three-dimensional electrode material [58,67], such that not all of the internal electrode surface is active for either electro-separation and/or electro-conversion. Then, the material thickness needs to be optimized, since the potential drop within the electrode increases when the thickness is increased [59]. Moreover, porous materials induce curvature effects on the electrical double-layer that can affect electro-separation processes [14,68–70]. These are important features to consider in process efficiencies, that are usually neglected in models and during optimization.

Another important feature to take into account in electrochemical systems applied for wastewater treatment is the mineral scaling issue due to electroprecipitation of hydroxides and calcium carbonate on the cathode (section 2.4) [22,47]. Knowing that Mg^{2+}, Ca^{2+} and CO_3^{2-} are ubiquitous in water, the progressive passivation of the cathode is almost unavoidable, especially for long-term electrolysis run in industrial applications [22]. Practically, the polarity is frequently reversed in order to redissolve the precipitates in bulk solution [51,71]. Under these conditions, both the cathode and the anode are typically the same materials in order to enable the electroprocesses to occur continuously, without interruption. It further means that the materials need to have the ability to be operated over a wide potential window without any oxidation or reduction that could modify their properties or even damage them. This symmetric constraint is often dismissed, because most of the research studies do not go beyond the laboratory scale with synthetic aqueous effluents. Considering the implementation of hybrid

electro-separation/electro-conversion systems operated simultaneously, this constraint cannot be met. The involvement of multiple electroprocesses requires an asymmetric system, since each electroprocess from section 2 imposes different requirements. Therefore, the cathode and anode materials have to be different. Moreover, there are conflicts in operating conditions (e.g., electrosorption/electrooxidation) and the degree of complexity increases with the number of electroprocesses involved.

3.2. A few recommendations and some emerging reactor designs

The interest in microreactors is gaining traction for wastewater treatment, due to the intensification of mass transfer and redox cycles, the ability to work with low conductivity solutions without adding supporting electrolyte, and the decrease in energy requirements [51,72–78]. However, the treatment capacity remains low with such devices, as they require a prefiltration step and do not solve the antagonist requirements issue when multiple electroprocesses are implemented.

Recently, the possibility to combine multiple electroprocesses in a single reactor has been proposed, by implementing the reactive electro-mixing concept and design [79]. In this setup, liquid or semi-liquid mixtures are mixed by the electrochemical (micro)-cells in movement connected in a monopolar mode, namely reactive electroblades. Therefore, it permits the combination of multiple (micro)-cells in one macro-reactor. Preliminary results have shown a 1.3-fold increase in paracetamol removal efficiency from wastewater by advanced electrooxidation with BDD, while the energy consumption was reduced by 20-fold compared to a conventional static filter-press cell involving the same anode material [79]. Following this approach, a single electroblade in rotation has been proposed using solid polymer electrolyte to operate in an even lower conductivity solution (μS cm^{-1} range) [80].

In addition, the possibility of bipolar mode in a static or electroblade hybrid system could be explored more for wastewater treatment applications, when different cathode and/or anode materials are required.

4. Conclusions and future perspectives

This short review highlights the synergies that could be obtained by combining electro-separation with electro-conversion simultaneously in a single hybrid reactor for wastewater treatment, reuse and recovery. However, conflicts in the constraints on the individual operations enforce the need to invent new designs and to improve the properties of reactive conductive materials for better selectivity and life span.

Flexible designs are needed to overcome the antagonist requirements of the different processes. The reactive electro-mixing reactor could be one of the solutions, but improvements are needed to increase electroprocess efficiencies and their number.

The theory of the electroporcess operations also needs to be revised, since both mass transport and reactions are involved in multiphasic and multiscale hybrid systems using porous polarized materials being subjected to angular momentum in some emerging reactor designs [81]. Mathematical models should be developed to predict the influence of operating parameters on the efficiency of different processes. Particular attention should be given to ensure symmetric modeling studies at the electrode/electrolyte interface, meaning that the degree of complexity and the scale level should be similar for both the electrode surface and in the electrolyte [82]. Coupling density functional theory (DFT) with molecular dynamics and microkinetics could be one of the possible approaches [82,83].

Finally, electrosynthesis as an electro-conversion system is attracting interest for the generation and subsequent valorization of value-added compounds from wastewater [16]. Moreover, H_2 electrogenerated at the cathode due to H_2O reduction could be produced in sufficient quantities at the industrial scale to be viable economically [84]. It could be even valorized as green H_2 if renewable energies are used to power the electrolyzer [85,86]. The possible synergy of co-electrogeneration (e.g., H_2, value-added compounds) with one or several electro-separation mechanisms should be carried out in the future, particularly with increases in selectivity. The efficiency of these processes remains to be tested with real wastewater effluents.

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