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Review

Electrocatalytic Oxidation: Enhancement Strategies for Catalyst Stability and Process Performance in Wastewater Treatment

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Abstract: Electrocatalytic oxidation (EO) is considered an effective technology for removing recalcitrant organic compounds from industrial wastewater; however, its practical deployment remains limited by electrode deactivation, high energy consumption, and insufficient performance under complex wastewater conditions. This review discusses recent advances in overcoming the intrinsic trade-off between oxidation activity, electrode durability, energy efficiency, and practical applicability of EO systems. Strategies ranging from heteroatom-based electrode engineering and process intensification to sustainable integration and data-driven optimization are critically summarized. Particular emphasis is placed on how doping strategies regulate electrode structures and reaction pathways, how process integration improves treatment efficiency, and how artificial intelligence enables predictive design and adaptive operation. Future efforts should emphasize durability assessment, real wastewater validation, and intelligent EO development to promote the practical implementation of EO in sustainable wastewater treatment.

Keywords: electrocatalytic oxidation; heteroatom doping; electrode stability; machine-learning; circular economy; wastewater treatment

1. Introduction

The poor amenability of organic pollutants in textile, pharmaceutical, petrochemical, and food industry wastewaters to conventional biological treatment makes electrocatalytic oxidation (EO) a promising solution [1,2]. Different from oxidation processes requiring external oxidants, EO produces highly reactive oxygen species (ROS), especially hydroxyl radicals (•OH), directly on the anode surface. These species can degrade a wide range of refractory organic pollutants and may achieve partial or complete mineralization into CO_2, H_2O, and inorganic ions under suitable conditions [3].

Despite these advantages, the practical application of EO is still restricted by several challenges. Electrode durability is one of the major limitations, as conventional Sb-SnO_2/Ti electrodes usually exhibit accelerated service lifetimes of only several hours at 100 mA/cm^2 in acidic sulfate electrolytes before obvious activity deterioration occurs [4–6]. In actual industrial wastewater treatment, energy consumption can reach tens of kWh/m^3, while current efficiency may decrease considerably in complex wastewater matrices, depending on wastewater characteristics, organic loading, reactor configuration, and the required treatment endpoint [7].

Compared with other advanced oxidation processes (AOPs), including ozonation and Fenton oxidation, EO provides several advantages, such as operation under ambient conditions, no requirement for additional chemical oxidants, and compatibility with renewable electricity, which facilitates automation and integration with existing treatment systems [8,9]. However, EO may require substantial electrical energy input and is also affected by electrode deactivation, whereas conventional homogeneous Fenton processes generate iron-rich sludge that requires

subsequent separation, treatment, and disposal [10]. Although the operating cost varies substantially with wastewater composition, electrode material, current density, and treatment endpoint, the relatively high electricity demand of EO may result in considerable operating costs compared with some conventional AOPs [11,12]. Therefore, improving electrode lifetime and reducing energy consumption remain essential for advancing EO toward practical applications.

Although numerous electrode materials have been developed over the past decades, no single anode simultaneously satisfies the requirements of high oxidation activity, long-term stability, low energy consumption, and economic feasibility under practical wastewater conditions. Consequently, recent research has gradually shifted from simply improving pollutant degradation efficiency toward rationally balancing catalytic activity, electrode durability, and process sustainability. This transition has stimulated the rapid development of heteroatom doping strategies, process intensification, hybrid treatment systems, and intelligent catalyst design.

Because anodic reactions determine both oxidant production and electrode degradation behavior, the development of advanced anode materials has become a key research direction in EO. An effective anode should exhibit high oxidant-generation ability, efficient formation of ROS and/or electrogenerated oxidants, sufficient electrochemical surface area while maintaining structural integrity, high oxygen evolution reaction (OER) overpotential, strong resistance to corrosion, passivation, coating delamination, and reactive-species attack, together with high electrical conductivity to reduce ohmic losses [13].

Based on their oxidation characteristics, EO anodes are generally divided into non-active and active anodes. Non-active anodes, including boron-doped diamond (BDD), PbO_2, Sb-doped SnO_2, and Magnéli-phase titanium suboxides (Ti_4O_7), usually exhibit high OER overpotentials and promote the formation of weakly adsorbed •OH, favoring extensive mineralization of organic pollutants. In contrast, active anodes, such as mixed metal oxide dimensionally stable anodes (DSA/MMO) and Pt-based electrodes, interact more strongly with surface oxygen species and commonly promote mediated oxidation pathways and/or active chlorine reactions, providing higher selectivity and structural stability [14].

The anodic potential drives pollutant degradation through direct and indirect oxidation pathways [10]. Direct oxidation involves electron transfer between pollutants and the electrode surface or reactions with adsorbed oxygen species generated during water oxidation. Indirect oxidation relies on electrochemically generated oxidants or reactive intermediates that diffuse into the solution and react with pollutants away from the electrode surface. The contribution of these two pathways is strongly influenced by electrode composition, electrolyte properties, pollutant characteristics, and operating conditions.

Among the challenges associated with EO, insufficient electrode stability remains a primary barrier for long-term operation. Therefore, extensive efforts have been directed toward improving electrode durability through heteroatom doping strategies [15]. Heteroatom doping introduces foreign elements, including metal ions or non-metal atoms, into electrode structures to modify electronic configurations, strengthen metal-oxygen interactions, inhibit structural transformation and corrosion, and regulate the adsorption and activation of reaction intermediates [16]. In addition, doping can enlarge electrochemically active surface areas and generate additional catalytic sites.

Besides electrode instability, the practical performance of EO is also constrained by inefficient electrical energy utilization, particularly when treating low-concentration pollutants. Moreover, oxidation reactions are often restricted by mass-transfer limitations because pollutants must diffuse from the bulk solution to the electrode surface where electrochemical reactions occur. These limitations can result in reduced current efficiency during practical operation. Overcoming these challenges requires not only durable electrode development but also improvements in reactor design, hybrid treatment approaches, and sustainable process integration. Accordingly, this review discusses recent advances in heteroatom doping for enhancing electrode stability, summarizes emerging approaches for improving EO process performance, and provides future perspectives for the practical implementation of EO in sustainable wastewater treatment.

2. Doping Strategies for Electrode Engineering

The practical application of electrocatalytic oxidation is mainly limited by electrode degradation, insufficient durability, and high energy demand during operation. Heteroatom doping provides an effective approach to regulate electrode structures, electronic properties, and surface reaction pathways, thereby improving catalytic activity and long-term stability. Figure 1 summarizes the degradation challenges of conventional anodes and illustrates how different heteroatom doping strategies modulate electrochemical properties toward enhanced electrocatalytic oxidation performance.

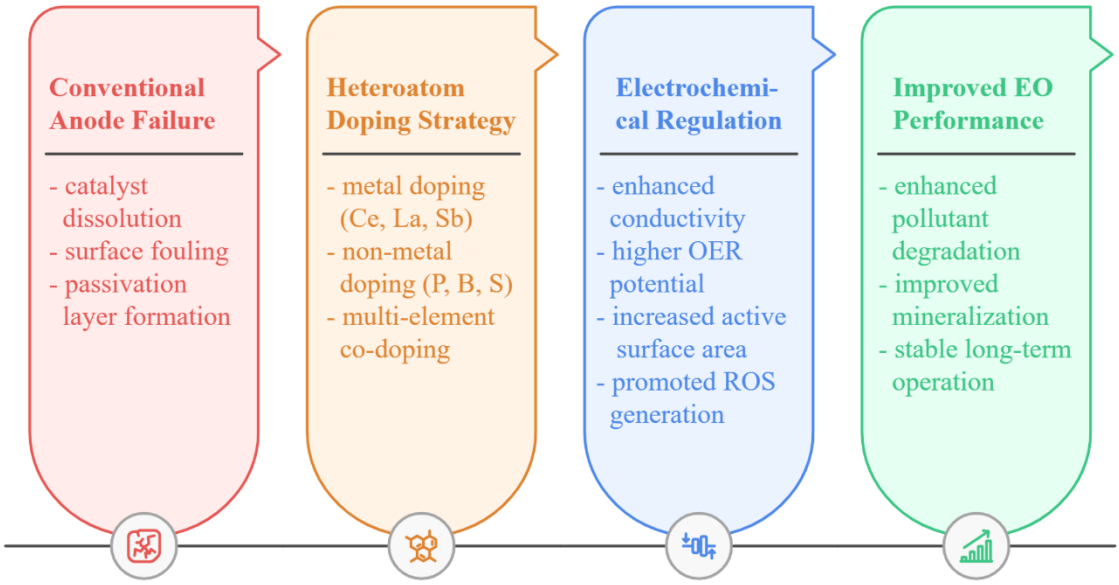

Figure 1. Schematic illustration of heteroatom doping strategies for regulating electrode properties and enhancing EO performance.

2.1. Single-Element Doping

Single-element doping introduces one foreign element into the electrode matrix to selectively regulate specific properties, including lattice stability, surface activity, electronic structure, and resistance toward passivation. Although this strategy generally improves one dominant limitation of conventional electrodes, its ability to simultaneously address multiple degradation mechanisms remains restricted.

Rare-earth elements are effective in improving the structural stability of PbO_2-based anodes. For example, Li et al. [17] reported that a Ce-doped TiO_2-NTs/PbO_2 anode exhibited an approximately 2.3-fold longer accelerated service life and a higher oxygen evolution potential than the corresponding Ce-free TiO_2-NTs/PbO_2 anode, demonstrating improved electrochemical durability and suppressed competitive oxygen evolution.

In addition to metal dopants, non-metal elements can also modify the electronic properties and catalytic behavior of electrode materials. Chille et al. [18] reported that phosphorus doping of ordered mesoporous carbons resulted in 53% phenol conversion and 40% benzoquinone (BQ) selectivity, which was higher than that of undoped ordered mesoporous carbons (OMC). The improved performance was mainly associated with P-induced electronic structure adjustment, the formation of additional defect sites, and enhanced exposure of catalytic regions. Unlike metal oxide modification, phosphorus doping in carbon materials primarily affects surface electronic properties and catalytic reactions rather than improving lattice stability.

2.2. Multi-Element Doping

Multi-element doping introduces two or more dopants with complementary functions to overcome the limitations of single-element modification. By simultaneously regulating electrode structures, electronic properties, and surface reaction pathways, multi-element doping provides a potential route to achieve a better balance between oxidation activity and long-term stability.

Co-doping with two different metals can tackle multiple failure mechanisms together. The synergistic interaction between different metal dopants can regulate lattice distortion, oxygen vacancy formation, charge-transfer behavior, and adsorption properties of reactive intermediates.

Yan et al. [19] introduced La and Ce into PbO_2 and developed a La-Ce-PbO_2-PVDF electrode. The modified electrode promoted hydroxyl radical production, exhibited a higher oxygen evolution potential (1.73 V), and showed a 3.12-fold larger electrochemically active surface area than the undoped anode. The co-doped electrode also improved methylene blue (MB) degradation and COD removal.

In addition to metal–metal systems, metal–non-metal and non-metal-non-metal co-doping strategies have also been explored in electrocatalytic systems because of their ability to induce electronic redistribution, defect engineering, and modified adsorption behaviors. For example, Fe/F co-doped CoO nanoneedles, Fe/N co-doped $CoSe_2$, B/P co-doped Pd nanostructures, and N/S co-doped carbon materials have demonstrated enhanced electrocatalytic activity through synergistic electronic effects [20,21]. However, many of these studies are still

performed in model electrochemical systems, and their direct applicability to practical EO wastewater treatment requires further validation.

As shown in Table 1, multi-element doping generally provides additional benefits compared with single-element modification by simultaneously regulating structural stability and catalytic properties. Nevertheless, the effectiveness of multi-element doping depends strongly on electrode composition, dopant interactions, operating conditions, and wastewater characteristics rather than representing a universal improvement.

Table 1. Representative doping strategies and modified anodes in electrocatalytic oxidation in wastewater treatment.

Anode Material	Dopants and Content	Key Indicators	Primary Active Species	Target Pollutants and Performance	Lifetime and Stability	Key Mechanism	Reference
TiO_2-NTs/Ce-PbO_2	Ce^{3+} (3 mM)	Oxygen evolution overpotential (1.81 V)	•OH, active chlorine Species	Rhodamine B (nearly complete removal)	139 h	Stress reduction, dense structure, indirect oxidation	[17]
OMCs	P (6 wt%); B (2 wt%); N (3 wt%)	Enhanced defect sites	•OH/Surface complexes	P-OMC: Phenol (53% conversion, 40% BQ selectivity)	/	Electronic modulation, defect engineering	[18]
La-Ce-PbO_2-PVDF	La, Ce	OER 1.73 V, 3.12× active surface area	•OH	Methylene Blue & COD removal	Significantly enhanced	Synergistic lattice protection, reduced leaching	[19]
SnO_2-based	Sb and other inorganic/metallic/non-metallic/rare-earth dopants	Oxygen evolution potential, electrochemical active surface area, conductivity, etc.	•OH	Various persistent organic pollutants (POPs), COD removal	Generally improve stability and service life	Electronic-structure regulation, oxygen-vacancy formation, enhanced charge transfer and ROS generation	[6]
PbO_2, RuO_2-IrO_2, DSA electrodes	Graphene nanosheets	Degradation efficiency, mineralization rate	•OH	Antibiotics and dyes	Improved long-term stability	Direct electron transfer and indirect oxidation mechanisms	[14]

2.3. Matrix Effects and Degradation

Although doping strategies have shown significant improvements under controlled laboratory conditions, their long-term performance still needs to be confirmed in practical wastewater systems. Most laboratory studies are conducted using simplified electrolyte solutions, whereas industrial wastewater contains complex components that can strongly affect oxidation pathways and electrode durability.

Constituents such as chloride (Cl^-), sulfate (SO_4^{2-}), bicarbonate (HCO_3^-), natural organic matter (NOM), suspended solids, and hardness ions (Ca^{2+}, Mg^{2+}) significantly alter oxidant speciation, promote byproduct formation, and accelerate electrode deactivation [6].

For example, chloride ions can promote the formation of active chlorine species and contribute to indirect oxidation; however, they may also cause pitting corrosion of titanium substrates and increase the risk of chlorinated byproduct formation [10]. Bicarbonate ions and NOM can consume hydroxyl radicals, resulting in decreased oxidation efficiency. Meanwhile, hardness ions and suspended particles may induce inorganic scaling, such as $CaCO_3$ and $Mg(OH)_2$ deposition, due to local pH changes near the anode surface. These deposits can hinder mass transfer and accelerate electrode passivation. Therefore, high activity obtained under ideal laboratory conditions does not necessarily indicate reliable performance during long-term wastewater treatment.

Under realistic operating conditions, complex wastewater matrices can severely accelerate specific anode failure mechanisms, including coating delamination, substrate corrosion via uncontrolled TiO_x interlayer growth, surface passivation, metal leaching (such as hazardous metal dissolution from coatings), and the persistent deposition of organic pollutants or inorganic scales. Although many doping strategies have achieved improved pollutant removal in short-term batch experiments, their actual contribution to extending electrode lifetime requires further assessment through standardized accelerated lifetime tests (ALT), continuous-flow reactors, and experiments using authentic wastewater rather than only synthetic systems.

3. Process Intensification and Hybrid Strategies

3.1. Advanced Electrode Materials

Emerging electrode materials provide new opportunities for developing EO anodes with improved oxidation capability, corrosion resistance, and structural stability for selective pollutant degradation. Medium-entropy oxides (typically containing three to four principal cations) and high-entropy oxides (containing five or more principal cations in near-equimolar ratios) possess highly disordered structures stabilized by configurational entropy. This entropy-driven stabilization can effectively retain active components, reduce metal dissolution (such as Pb leaching from PbO_2 coatings), and inhibit the formation of passivation layers under strong anodic polarization [22]. However, their long-term behavior under realistic EO wastewater conditions remains to be systematically evaluated.

Metal-organic framework (MOF)-derived catalysts represent another promising class of EO electrode materials. Through pyrolysis of MOF precursors, hierarchical carbon structures containing highly dispersed metal active sites can be obtained. The combination of conductive carbon frameworks and uniformly distributed metal sites provides abundant electrochemically active regions and interconnected pore structures, which can facilitate pollutant adsorption and alleviate mass-transfer limitations during EO treatment [23,24].

3.2. Machine Learning-Guided Catalyst Design

The traditional trial-and-error approach for optimizing dopant combinations and electrode compositions is often inefficient and requires extensive experimental resources. Machine learning (ML) provides a data-driven alternative by establishing relationships between material characteristics, operating parameters, and EO performance. An effective ML framework for EO applications should integrate both material-related descriptors and process-level variables. These inputs may include elemental composition, morphology, work function, surface roughness, pollutant type, TOC/COD concentration, pH, current density, mass-transfer conditions, energy consumption, byproduct generation, and electrode lifetime [25,26]. Furthermore, active learning strategies, in which ML models identify the most informative experiments for subsequent validation, offer a promising route to reduce unnecessary experimental screening [27].

3.3. Electrode Regeneration Strategies

Even highly optimized doped electrodes gradually lose activity during prolonged operation because of surface fouling, catalyst dissolution, and structural degradation [28]. Therefore, in situ regeneration methods have been explored to recover electrode activity and extend operational lifetime. Approaches including periodic polarity reversal, pulsed current operation, dilute acid cleaning, and electrochemical annealing have been adopted from other electrochemical systems. However, their effectiveness in EO wastewater treatment requires further investigation because electrode deactivation is strongly influenced by complex factors such as organic deposition, inorganic scaling, salinity, and pH fluctuations. For example, polarity reversal may remove weakly attached foulants from electrode surfaces, whereas pulsed current operation can reduce concentration polarization. Nevertheless, optimized regeneration protocols under realistic wastewater conditions remain insufficiently established and require long-term evaluation in continuous-flow systems.

3.4. Hybrid Treatment Processes

EO is effective when combined with other treatments. Electrocatalysis combined with biological treatment can serve as a pretreatment step to break down recalcitrant compounds into more biodegradable intermediates [2,29].

Guo et al. [30] degraded pharmaceuticals and personal care products (PPCPs) from wastewater using electrocatalysis and biological treatments. Removals of 71.91% of diclofenac at 0.73 mA/cm^2 and 58.92% of clofibric acid at 0.64 mA/cm^2 were observed, respectively, which were 42.79% and 17.52% higher than the biological treatment alone. Electrocatalysis can increase the biodegradability of substances, making it easily removed in the subsequent biological stage.

Electrochemical pretreatment followed by membrane filtration has been explored for enhanced pollutant removal and water reuse. For example, Meiramkulova et al. [31] used electrochemical (EC) pretreatment to treat slaughterhouse wastewater, in which the EC pretreatment unit removed 71–85% of the major pollutants but poorly removed chlorine, nitrites, nitrates, phosphates, total phosphorus, and ammonium nitrogen. The subsequent membrane filtration further improved overall pollutant removal, while the electrochemical pretreatment also reduced membrane cake formation.

Electrocatalysis combined with adsorption allows high-concentration pollutants to be first adsorbed onto activated carbon, followed by electrocatalytic polishing of the remaining low-concentration contaminants.

Electrocatalysis coupled with electro-Fenton systems can generate hydroxyl radicals at the anode and hydrogen peroxide at the cathode, thereby enabling in situ Fenton chemistry [32].

4. Sustainability and Circular Economy Considerations

Electrocatalytic wastewater treatment should be evaluated from a circular economy perspective by considering resource recovery, electrode material recycling, and reduction of the overall environmental impact.

Organic pollutants can be partially or completely mineralized to CO_2 and H_2O, and inorganic species during EO treatment, depending on the operating conditions and pollutant characteristics. However, inorganic species, especially metals such as Cr, Cu, Ni and precious metals can be recovered from the concentrated retentate or spent electrolyte by cathodic electrodeposition. Spent electrodes contain valuable materials—titanium substrate, dopant metals (Ce, Bi, Sb), and in some cases, precious metals. The recovery and recycling of valuable components from spent electrodes provide opportunities to reduce material consumption and environmental impacts [33]. However, comprehensive studies are still needed to optimize extraction efficiencies across varied electrode formulations and operational leaching conditions.

Life cycle assessment (LCA) of electrocatalytic treatment should include energy consumption and production, and the transport of electrode materials; the frequency of electrode replacement; and the disposal or recycling of spent electrodes, as well as chemical consumption for pH adjustment or electrolyte maintenance [34].

Renewable energy integration should also be considered because electrocatalytic treatment is inherently electricity-dependent. Coupling electrochemical oxidation systems with renewable energy sources, such as photovoltaic or wind power, can reduce reliance on grid electricity and lower the associated environmental impacts [35]. Previous studies have demonstrated the technical feasibility of directly powering electrochemical wastewater treatment systems using photovoltaic panels or wind turbines [36,37]. However, fluctuations in renewable electricity supply may influence treatment performance, highlighting the importance of appropriate power-management and energy-storage strategies [38].

5. Future Perspectives

Although considerable progress has been made in improving electrode stability and EO process performance, the translation of laboratory-scale achievements into practical wastewater treatment remains challenging. Many existing studies are based on short-term degradation tests using synthetic solutions, whereas real applications require reliable operation in complex wastewater matrices, comparable performance evaluation, and economically viable process configurations. Future efforts should therefore focus not only on improving individual performance metrics but also on developing standardized assessment methods, realistic validation platforms, and intelligent operating strategies (Figure 2).

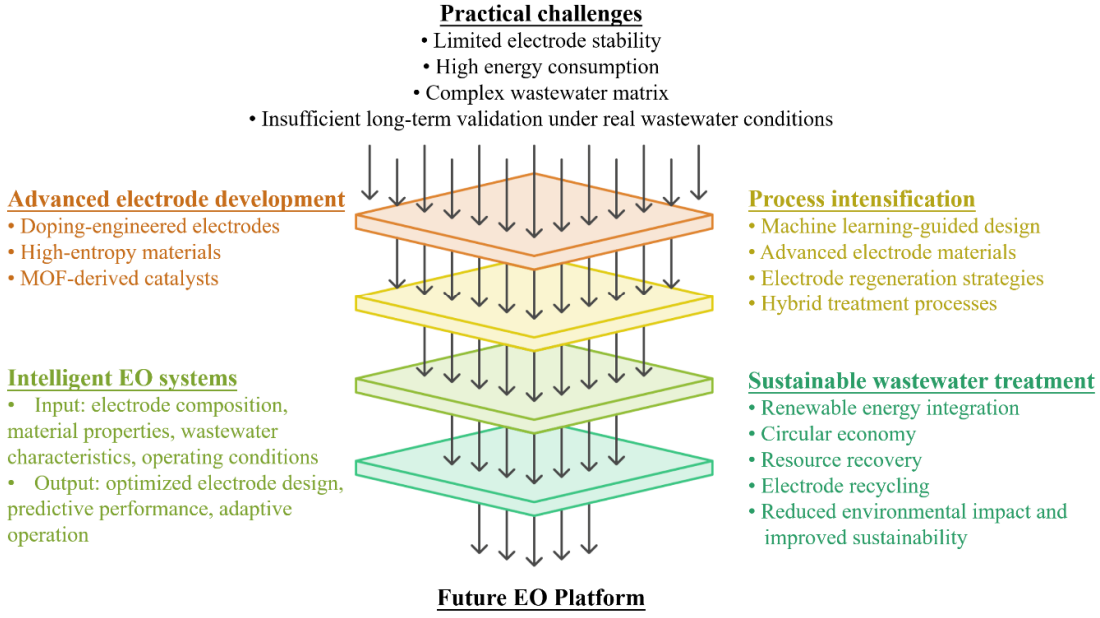

Figure 2. Integrated strategies toward practical EO for sustainable wastewater treatment.

5.1. Standardized Evaluation of Electrode Performance

The absence of consistent evaluation criteria remains a major obstacle when comparing different EO electrode systems. Reported removal efficiencies are often obtained under diverse operating conditions, including different current densities, electrolyte compositions, reactor designs, and reaction times, making direct comparison among studies difficult.

Future studies should adopt more systematic durability assessments, including accelerated lifetime testing (ALT), long-term continuous-flow operation, energy consumption evaluation, and detailed analysis of electrode degradation mechanisms. Electrode lifetime should be assessed not only by maintaining pollutant removal efficiency but also by monitoring changes in surface structure, catalyst dissolution, coating adhesion, and electrochemical properties during extended operation.

Establishing reliable benchmarking protocols will improve the comparability of emerging electrode materials and support the rational selection of EO systems for specific wastewater applications.

5.2. Bridging Laboratory Studies and Real-World Applications

The gap between laboratory-scale investigations and industrial wastewater treatment remains a critical barrier to EO commercialization. Many newly developed electrode materials exhibit excellent performance in simplified electrolyte systems but experience decreased activity and accelerated degradation when exposed to real wastewater containing chloride, natural organic matter, suspended particles, and inorganic ions [39].

Future research should place greater emphasis on validation using authentic wastewater, pilot-scale continuous-flow reactors, and long-term operation under realistic conditions. In parallel, interactions between electrode materials and complex wastewater components should be systematically examined to understand degradation pathways and guide the design of more durable electrodes.

Furthermore, techno-economic analysis should be incorporated into electrode and process development, because high pollutant removal efficiency alone cannot ensure practical feasibility without considering electricity demand, electrode replacement frequency, and overall operational costs.

5.3. Intelligent and Adaptive EO Systems

Beyond improvements in materials and process design, future EO technologies are expected to advance toward intelligent treatment platforms that combine data-driven prediction, automated optimization, and adaptive operation [26]. ML approaches provide new opportunities to accelerate electrode discovery and process optimization by establishing quantitative relationships between electrode composition, structural characteristics, operating parameters, and treatment performance.

An important future direction is the construction of integrated ML platforms that can predict suitable electrode compositions, dopant combinations, synthesis parameters, and operating conditions according to specific targets, such as oxidation efficiency, energy consumption, and electrode lifetime. However, the application of ML in EO is currently restricted by limited datasets and inconsistent experimental conditions, which reduce model reliability and generalization ability.

Future EO databases should incorporate multidimensional descriptors rather than relying only on elemental composition. These descriptors should include morphological properties (e.g., grain size, surface roughness, and porosity), electronic characteristics (e.g., work function, electronic structure, and density of states), as well as engineering parameters including pollutant properties, COD/TOC concentration, pH, current density, mass-transfer conditions, energy consumption, byproduct formation, and electrode lifetime. Such comprehensive datasets would allow ML models to better capture the connections between material structure, electrochemical behavior, and practical treatment performance, enabling mechanism-informed catalyst design instead of simple composition screening.

Transfer learning may provide an effective solution to the data scarcity problem in EO systems. Knowledge obtained from related electrocatalytic fields, such as water splitting and CO_2 reduction, could provide transferable information regarding metal–adsorbate interactions, electronic structure–activity relationships, and electrochemical reaction trends. By pre-training models using these larger datasets and subsequently adapting them with EO-specific data, the amount of experimental data required for reliable prediction may be significantly reduced.

In addition, uncertainty evaluation should become an essential component of ML-assisted EO design. Instead of providing only single-value predictions, probabilistic models, ensemble learning, and Bayesian approaches can estimate prediction confidence and identify uncertain regions in the material and operating parameter space. These high-uncertainty regions may represent unexplored opportunities for discovering new electrode systems, allowing experimental efforts to be directed toward promising candidates while avoiding unreliable predictions.

Ultimately, integrating ML, standardized experimental databases, and automated electrochemical platforms may transform EO development from empirical optimization toward intelligent systems capable of predictive design, adaptive operation, and accelerated electrode discovery.

6. Conclusions

EO has emerged as a promising technology for treating recalcitrant wastewater contaminants, yet its practical deployment remains limited by the intrinsic trade-off among oxidation activity, electrode durability, energy efficiency, and operational stability. This review highlights that overcoming these challenges requires a transition from isolated catalyst optimization toward integrated electrode–process engineering. Heteroatom doping provides an effective approach to regulate electrode structures and reaction pathways, while multi-element strategies offer opportunities for synergistic enhancement of activity and stability. The combination of process intensification, sustainable design, and intelligent technologies offers new opportunities for improving EO efficiency and scalability. Further development will rely on standardized performance evaluation, validation under real wastewater conditions, and data-driven approaches that enable predictive design and adaptive operation. By integrating materials innovation, process engineering, and artificial intelligence, next-generation EO systems may evolve from empirical development toward more efficient, durable, and scalable wastewater treatment technologies.

Author Contributions

X.C.: methodology, writing—original draft preparation, data curation, visualization, investigation; D.-J.L.: conceptualization, writing—reviewing and editing, supervision, funding acquisition, project administration. All authors have read and agreed to the published version of the manuscript.

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Use of AI and AI-Assisted Technologies

The AI tool was used to polish the texts and presentations.

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