Decoupling mass transport from spin chemistry in electro-Fenton process under magnetic fields

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Supplementary Tables

Table S1. Timeline of representative academic developments directly related to electro-Fenton mechanisms, magnetic-field-assisted electrochemistry, and spin-chemistry regulation.

Period / Year	Representative research stage	Main research focus	Relevance to this review	References
2009	Classical electro-Fenton mechanism	Cathodic H_2O_2 generation, Fe^{2+}/Fe^{3+} cycling, •OH production, pollutant degradation kinetics, and pH-dependent Fenton chemistry	Established the fundamental EF reaction network and clarified the central roles of H_2O_2, Fe^{2+}/Fe^{3+}, H^+, and •OH in pollutant degradation	错误!不能识别的开关参数。错误!未找到引用源。
2016	Magnetic regulation of oxygen reduction on ferromagnetic catalysts	Internal magnetism of ferromagnetic catalysts, external magnetic-field response, and ORR activity enhancement	Provided early evidence that catalyst magnetism can influence oxygen-related electrochemical reactions, offering a background for discussing magnetic effects on O_2 activation in EF	错误!不能识别的开关参数。
2020	Radical-pair spin regulation in electrocatalytic reactions	Magnetic-field regulation of radical-pair spin states, singlet–triplet interconversion, and spin-dependent reaction pathways	Provided an electrocatalytic example showing that magnetic fields can influence radical-pair spin evolution and reaction selectivity	错误!不能识别的开关参数。错误!未找到引用源。
2022	Magnetic-field-assisted electrochemistry	Lorentz force, Kelvin force, MHD convection, micro-MHD effects, interfacial mass transport, and magnetic-field-induced concentration redistribution	Provided the physical basis for understanding how magnetic fields regulate mass transport, diffusion layers, bubble behavior, and interfacial reactant enrichment	错误!不能识别的开关参数。
2023	Fe^{2+} regeneration and EF rate-limiting steps	Fe^{3+} reduction, Fe^{2+} regeneration, electron-transfer pathways, mass-transport limitations, and catalyst-assisted iron cycling	Clarified that Fe^{2+} regeneration is a key bottleneck in EF and provided the mechanistic basis for analyzing magnetic-field-enhanced Fe^{2+}/Fe^{3+} cycling	错误!不能识别的开关参数。错误!未找到引用源。

Period / Year	Representative research stage	Main research focus	Relevance to this review	References
2024	Quantitative decoupling of magnetic-field effects	Separation of Lorentz-force-driven mass transport from intrinsic kinetic contributions using controlled magnetoelectrochemical systems	Provided an experimental strategy for distinguishing apparent activity enhancement caused by MHD convection from possible intrinsic spin-related kinetic effects	错误!不能识别的开关参数。
2025	Spin magnetic effects in oxygen electrocatalysis	Magnetic-field-induced spin regulation at metal sites, O–O intermediate formation, and spin-sensitive oxygen reaction pathways	Further supported the relevance of spin-state regulation in oxygen-related electrocatalysis, offering useful analogies for EF cathodic O_2 activation	错误!不能识别的开关参数。

Table S2. Representative examples of magnetic-field regulation of iron species and Fe^{2+}/Fe^{3+} cycling in EF-related systems.

Catalyst System / Electrode Material	**Magnetic Field Conditions**	**Mechanistic Explanation of Magnetic Field Influence**	**Reference**
Carbon felt cathode–magnetite nanoparticle composite	External magnetic bar to retain catalyst	Enhances retention of magnetic catalyst in electrode pores, improves Fe^{3+} to Fe^{2+} regeneration rate, boosts •OH generation, and increases EF efficiency.	[8]
RuO_2–IrO_2/Ti electrode–zero-valent iron	Magnetic ZVI decorated on electrode surface	Provides a continuous source of Fe^{2+}, promoting H_2O_2 decomposition and •OH generation, accelerating the EF process.	[9]
3D Electro-Fenton system–magnetic MWCNTs	Embedded magnetic MMWCNT particle electrodes	Magnetization of particle electrodes themselves enhances •OH production and boosts EF catalytic activity.	[10]
Charcoal-shaped $NiFe_2O_4/Fe_2O_3$ catalyst	Magnetic catalyst recovery / magnetic separation	Dissolved Fe^{2+}/Fe^{3+} and surface iron sites participate in Fenton-like reactions, enhancing RhB degradation	[11]
Magnetic-field-assisted EF process / electrocatalytic cathode	Static magnetic field-assisted mass transport	Promotes Fe^{3+} delivery to cathodic sites and supports $Fe^{3+} \rightarrow Fe^{2+}$ regeneration	[4]
Fe_3O_4 magnetic nanoparticles in EF	Magnetic iron oxide nanoparticles	Fe_3O_4 provides surface Fe sites and supports Fe^{2+}/Fe^{3+} cycling for pollutant degradation	[12]
Pd/Fe_3O_4 nanocomposite catalyst	Magnetically assisted catalyst loading and recovery	Pd promotes H_2O_2 generation, while Fe_3O_4 supplies iron active sites for EF degradation	[13]

Table S3. Research progress on the magnetic field regulation of iron ion activity.

Catalyst System / Electrode Material	Magnetic Field Conditions	Mechanistic Explanation of the Magnetic Field Influence	Reference
Fe^{2+}/H_2O_2 Fenton system for methyl blue degradation	Static magnetic field	Magnetic field enhances Fenton reaction efficiency and promotes dye degradation, mainly by improving Fe^{2+}/H_2O_2 reaction and •OH generation.	[14]
Reductive iron powder / H_2O_2 system	Weak magnetic field	Enhances Fe^0 corrosion, accelerates Fe^{2+} generation, produces more •OH radicals, and improves 4-nitrophenol degradation	[15]
Fe_3O_4 nanoparticles / H_2O_2 system	Alternating magnetic field	AMF accelerates free-radical generation by iron oxide nanoparticles, mainly through magnetothermal and field-induced effects	[16]
Zero-valent iron / H_2O_2 Fenton-like system	Static magnetic field	Enhances mass transport, modifies the surface oxide layer, shortens the lag phase, and promotes •OH generation for 4-chlorophenol degradation.	[17]
Zero-valent iron / As(III) system with coexisting anions	Weak magnetic field	Promotes ZVI corrosion and Fe species migration; coexisting anions participate in charge balance and affect Fe-mediated contaminant removal.	[18]

Zero-valent iron / H_2O_2 Fenton-like system for X-3B decolorization	Weak magnetic field	Improves Fe^0 corrosion, enhances Fe^{2+} release, increases dye decolorization efficiency, and broadens the effective pH range.	[19]
PVP/Fe_3O_4@SiO_2 nanoporous catalyst / sono-photo-Fenton system	Static magnetic field	Magnetic Fe_3O_4@SiO_2-based catalyst promotes Fe^{2+}/Fe^{3+} cycling and enhances Toluidine Blue degradation under coupled sono-photo-Fenton conditions.	[20]
FeCu/rGO photo-Fenton catalyst	Static magnetic field	Magnetic field enhances radical intensity and accelerates pollutant degradation by promoting interfacial Fe/Cu redox cycling and ROS generation.	[21]
Magnetic TiO_2/Fe_3O_4-chitosan microbeads / photo-electro-Fenton system	Static magnetic field	Magnetic Fe_3O_4 enables catalyst recovery and contributes to heterogeneous Fenton-like activity, although the direct magnetic-field effect on activity is limited.	[22]
Magnetite–silver composite / photo-Fenton system	Magnetic separation / magnetic response	Magnetite provides magnetic recoverability and Fe-based Fenton-like sites, while Ag improves charge separation and photo-Fenton activity.	[23]

Table S4. Evidence and analogies for magnetic-field regulation of proton transport, local pH, and interfacial water behavior in electrochemical systems.

System / Electrode Material	Magnetic Field Conditions	Relevance to magnetic-field-assisted EF	Reference
Co electrodeposition system	Static magnetic field	Directly supports the idea that magnetic fields can regulate interfacial pH polarization near electrodes, which is highly relevant to EF microenvironment control	[24]
Non-magnetic electrocatalytic electrodes	0.5–1 T static magnetic field	Supports the role of MHD convection in replenishing reactants and mitigating local concentration polarization, including possible H^+ depletion in EF	[6]
Enzymatic biofuel cell anode	Constant parallel magnetic field	Provides indirect evidence that magnetic fields may affect H_3O^+ transport in electrochemical microenvironments	[25]
PEM fuel cell cathode catalyst layer with magnetic particles	Constant magnetic field	Provides analogy for magnetic-field regulation of water/proton pathways, but not direct EF evidence	[26]
Magnetically aligned proton exchange membranes	0–1 T magnetic field	Useful analogy for proton-channel regulation, but mainly relevant to membrane systems rather than bulk EF electrolytes	[27]
Electrochemical double layer at magnetized interfaces	Static magnetic field	Provides supporting evidence that magnetic fields may alter interfacial proton-coupled electron-transfer distance, but the EF contribution remains to be verified	[28]

Table S5. Advanced operando characterization and theoretical calculation methods for validating magnetic-field-assisted electro-Fenton mechanisms.

Mechanism	Key information	Mechanism revealed in magnetic-field-assisted EF	Representative output	References
In situ/operando Raman and FTIR spectroscopy	Catalyst reconstruction, surface functional groups, adsorbed intermediates, and metal–oxygen bonding evolution	Can monitor catalyst structural changes and adsorbed oxygen-containing intermediates under electrochemical operation; useful for determining whether magnetic fields alter surface reconstruction or reaction pathways	Raman bands, FTIR absorption peaks, intermediate fingerprints	[29]
Operando XAS, including XANES and EXAFS	Metal oxidation state, local coordination structure, bond length, and dynamic active-site evolution	Can track Fe^{2+}/Fe^{3+} redox cycling and coordination changes during EF operation; useful for verifying whether magnetic fields accelerate iron cycling or stabilize active Fe sites	XANES edge shift, EXAFS fitting, coordination number, Fe–O bond distance	[30]
EPR/ESR spin-trapping spectroscopy	ROS species, radical signals, and possible spin-related intermediates	Can detect •OH, O_2•$^-$, $^1O_2$, and other spin-active species; useful for evaluating whether magnetic fields affect ROS generation or radical-pair evolution, but strict controls are needed to exclude MHD and pH effects	DMPO–•OH, DMPO–O_2•$^-$, TEMP–$^1O_2$ signals	[31]

Operando electrochemical techniques, including EIS, RRDE, DEMS, and EQCM	Charge-transfer resistance, H_2O_2 selectivity, gaseous products, mass variation, and interfacial kinetics	Can quantify charge-transfer resistance, H_2O_2 yield, gas evolution, and catalyst reconstruction; useful for separating electron-transfer changes from mass-transport enhancement	Nyquist plots, ring current, H_2O_2 Faradaic efficiency, mass-change curves	[32]
DFT and spin-polarized DFT calculations	Adsorption energy, electronic structure, spin density, charge redistribution, and reaction free-energy barriers	Can compare O_2, *OOH, H_2O_2, Fe species, and ROS-related intermediates on different catalyst surfaces; useful for evaluating possible spin-dependent reaction pathways	ΔE_{ads}, ΔG diagrams, DOS/PDOS, spin-density maps, Bader charge	[33]
AIMD and molecular simulations	Interfacial water structure, proton transfer, hydrogen-bond network dynamics, and solvation environment	Can explore proton transfer and water-network reorganization near the electrode interface; useful for testing whether magnetic-field-related solvent-structure hypotheses are plausible, but not direct proof of EF enhancement alone	Atomic trajectories, radial distribution functions, hydrogen-bond lifetime, proton-hopping pathways	[34]
Integrated operando reactor design and best-practice protocols	Compatibility between magnetic fields, electrochemical cells, optical/spectroscopic windows, and realistic reaction conditions	Can improve the reliability of mechanistic interpretation by ensuring that operando measurements are performed under controlled magnetic-field, hydrodynamic, and electrochemical conditions	Standardized cell design, control experiments, operando workflow	[35]

Table S6. Mechanisms and examples of magnetic-field regulation of oxygen-related catalytic reactions and their relevance to EF systems.

Catalyst System / Electrode Material	Magnetic Field Conditions	Mechanistic Explanation of Magnetic Field Influence	Reference
Photocatalytic systems	Electric or magnetic field-assisted photocatalysis	External fields may promote charge separation, carrier migration, and surface oxygen activation in photocatalytic reactions.	[36]
Non-magnetic electrocatalytic electrodes	Static magnetic field	Magnetic fields enhance ORR mainly by accelerating O_2 diffusion and mass transport.	[37]
CoPt alloys and Pt/FeCo heterostructures	External magnetic field and residual magnetism	Magnetic fields and catalyst magnetism accelerate O_2 diffusion and improve ORR activity.	[38]
Ferromagnetic catalysts	Constant magnetic field	Internal magnetism of ferromagnetic catalysts influences O_2 adsorption and ORR catalytic activity under external magnetic fields.	[2]
Fe-N-C single-atom catalyst	Crystal-field regulation combined with external magnetic field	Magnetic field and ligand-field modulation regulate Fe spin state, optimizing O_2 adsorption and *OOH formation energetics.	[39]

$Ni_5Fe_1(OH)_2$ (OER catalyst, analogous to ORR)	External magnetic field	Spin magnetic effect at Fe sites promotes O–O intermediate formation and oxygen evolution pathways.	[7]
Core-shell nanoparticle catalysts	Constant magnetic field	Magnetic fields modulate catalytic properties of core-shell particles, potentially affecting adsorption and desorption processes.	[40]
Electrocatalysts under alternating magnetic field	Alternating magnetic field	AMF may induce magnetothermal effects, eddy currents, and non-thermal magnetic effects that change catalytic activity.	[41]
Multiscale catalytic systems	Non-contact constant magnetic field	Magnetic fields may couple spin polarization, electromagnetic effects, and interfacial catalytic processes.	[42]
Multiferroic or magnetoelectric electrocatalytic systems	Constant magnetic field	Magnetoelectric coupling may modulate spin-related electronic structures and catalytic activity	[43]

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