

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

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ABSTRACT: Electro-Fenton (EF) is one of the most effective processes for treating organic pollutants in water. However, its performance is often restricted by mass transport limitations and slow catalytic reactions. While recent reviews have summarized various strategies to enhance the EF process performance, a focused analysis separating the physical magnetohydrodynamic (MHD) effect on mass transport from the quantum spin-chemistry effect on reaction kinetics—one of the most promising enhancement routes—has not yet been established. Here, this gap is addressed by developing a framework including the four key species of the EF systems: the $\text{Fe}^{2+}/\text{Fe}^{3+}$ redox couple, the reactants (H^+ and O_2), and the electrons supplied. On this basis, the influence of magnetic fields on each species is examined, clearly distinguishing MHD-driven mass transport from spin-chemistry effects on intrinsic kinetics. This distinction helps resolve ongoing mechanistic debates and provides practical guidance for designing advanced magnetically assisted water treatment technologies.

KEYWORDS: Advanced oxidation processes (AOPs), Electro-Fenton (EF), magnetic field, magnetohydrodynamics (MHD), mechanistic study, spin chemistry.

Highlight

- [1] A four-species framework decouples transport from spin chemistry in electro-Fenton.
- [2] $\text{Fe}^{2+}/\text{Fe}^{3+}$, H^+ , O_2 , and electrons anchor multiscale mechanistic analysis.
- [3] A quantitative strategy estimates mass-transfer and residual kinetic contributions.
- [4] Mechanistic decoupling guides operando validation and catalyst-reactor co-design.

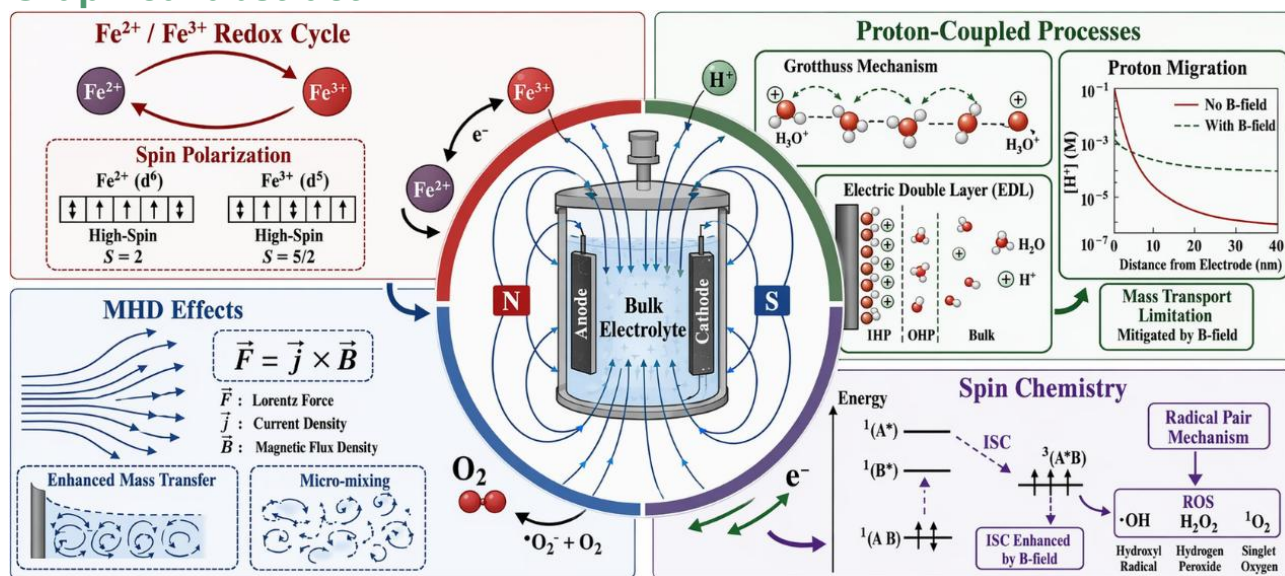
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Graphical abstract



1. Introduction

Water pollution by hard-to-degrade organic xenobiotics is a major global environmental problem nowadays. The urgency arising from this situation has driven the development of advanced oxidation processes (AOPs). Among them, the electro-Fenton (EF) process is especially promising because it generates powerful hydroxyl radicals ($\cdot\text{OH}$) *in-situ* under mild conditions^[1]. However, the efficiency of EF is still limited due to the low cathodic H_2O_2 yield^[2], the slow electrocatalytic Fe^{3+} reduction^[3], and its strong dependence on local proton (H^+) concentration^[4]. To keep iron soluble and promote $\cdot\text{OH}$ formation, the bulk pH must be kept strictly around 3.0; any deviation causes rapid precipitation of ferric hydroxide ($\text{Fe}(\text{OH})_3$) and a sharp loss of catalytic activity. External magnetic fields have been explored as a potentially useful non-contact approach to improve the EF performance^[5]. For example, our prior work showed that a magnetic field markedly boosted the electrocatalytic performance by tuning the e_g orbital electron occupancy of cobalt active sites^[6].

To further demonstrate the rapid development of this field, **Fig. 1** presents the annual publication number and citation counts related to magnetic-field-assisted electro-Fenton and magnetically regulated electrocatalysis from 2006 to 2026. As shown in Fig. 1a, the number of publications increased gradually before 2015 and then rose sharply after 2018, reaching more than 300 papers per year in recent years. Meanwhile, the citation counts also increased

continuously, from only a few citations in the early stage to more than 20,000 citations in 2025 (**Fig. 1b**). The relatively lower values in 2026 are mainly because the data for this year are still ongoing. This increasing trend indicates that magnetic-field regulation has attracted growing attention and has evolved from a simple auxiliary mass-transfer strategy to a broader research topic involving interfacial microenvironment regulation, catalyst design, and spin-dependent reaction kinetics. Therefore, a clearer mechanistic framework is needed to distinguish the different roles of magnetic fields in EF systems.

Magnetic fields affect the EF system in two main ways. The first is the so-called magnetohydrodynamic (MHD) effect, which arises from the Lorentz force exerted on charged species dissolved in the electrolyte; this causes convection that improves mass transport^[7], removes gas bubbles from the electrode surface, and helps maintain the electrode activity. The second phenomenon comprises the spin-chemistry effects, in which the magnetic field changes the spin states of paramagnetic species such as Fe^{2+} , Fe^{3+} , O_2 , and radicals; these effects can change the reaction kinetics and product selectivity by altering the rate of intersystem crossing^[8], modifying the energy barriers of spin-dependent reactions, or changing adsorption and catalysis on magnetic catalyst surfaces^[9].

To visualize this central idea, **Fig. 2** summarizes the conceptual framework of magnetic-field-assisted EF process. The four key species, namely the $\text{Fe}^{2+}/\text{Fe}^{3+}$ pair, H^+ , O_2 , and electrons, are arranged around the central EF reactor to show how magnetic fields

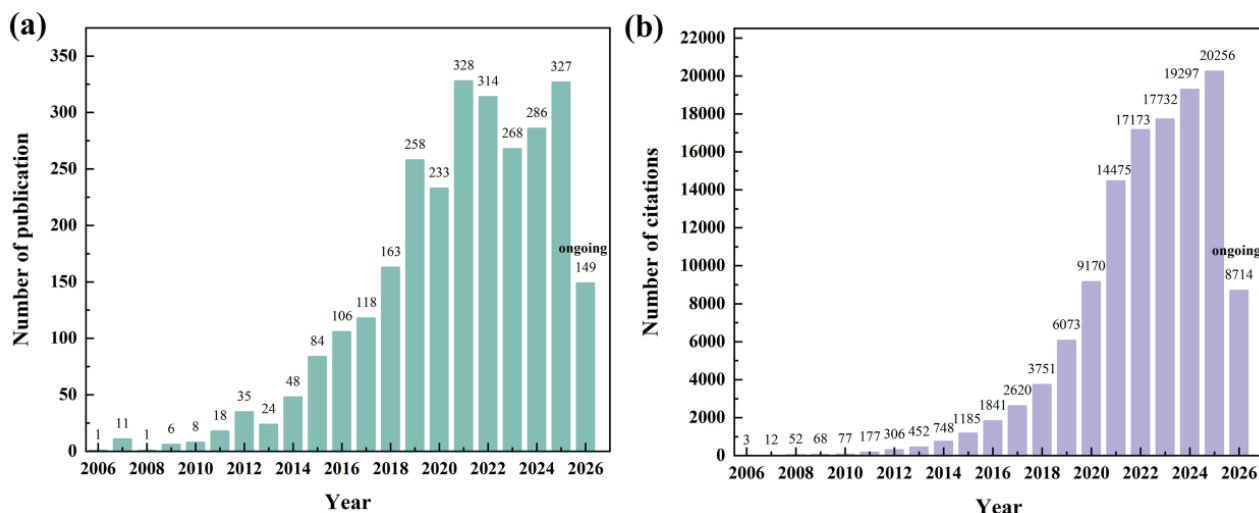


Fig. 1. Bibliometric analysis of magnetic-field-assisted electro-Fenton and magnetically regulated electrocatalysis. (a) Annual number of publications from 2006 to 2026. (b) Annual citation counts from 2006 to 2026. The data for 2026 are still ongoing and therefore do not represent a full-year trend.

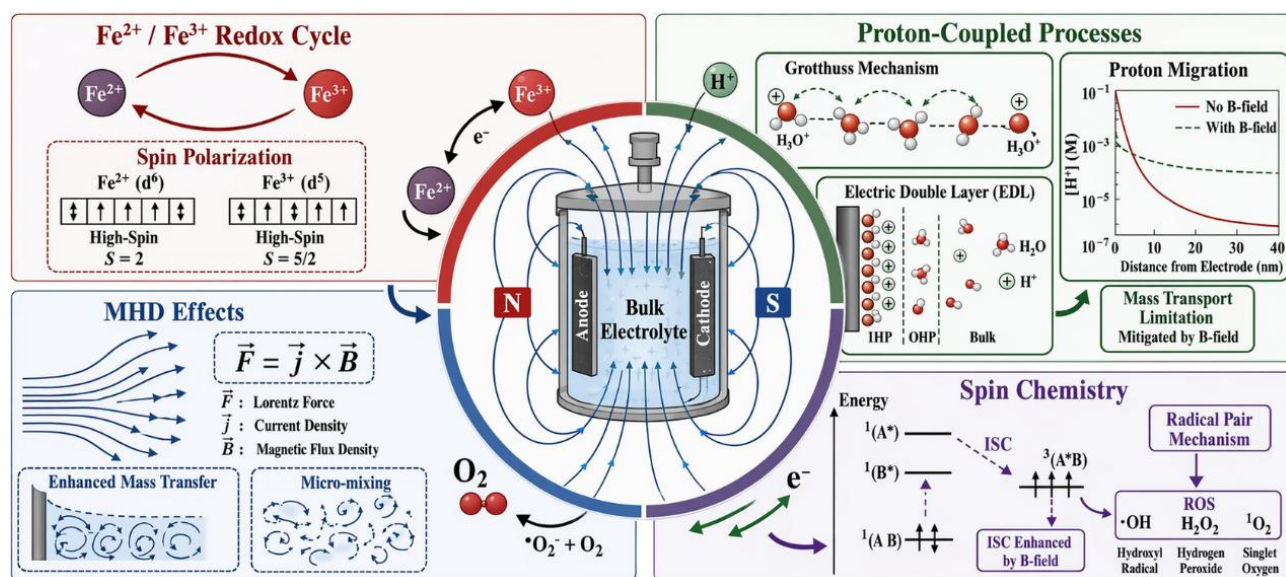


Fig. 2. Framework for separating mass transport from spin chemistry effects in magnetic field-assisted EF systems.

regulate the system through two coupled but distinguishable pathways. The left half represents physical MHD effects, including Lorentz-force-driven convection, bulk mixing, and ion transport, whereas the right half represents quantum spin-chemistry effects, including spin-state regulation, spin-dependent reaction kinetics, and O₂ activation toward H₂O₂ and $\cdot OH$ formation. This scheme establishes the main logic of this review: magnetic-field enhancement in EF must be analyzed by decoupling macroscopic mass transport from microscopic spin-

dependent kinetics.

A major challenge in this area arises from the fact that EF performance depends on both macroscopic fluid dynamics and quantum spin effects. Although many studies report better results when magnetic fields are applied [10], the specific underlying mechanisms are still unclear. The main questions are: how much of the improvement is associated with the enhanced mass transport [11], and how much to changes in reaction kinetics [12]? Do these effects act mainly on individual species or through their

interactions ^[13]? And what role magnetic materials play ^[14, 15]? To improve readability and provide a clearer historical background, Table S1 in the Supporting Information summarizes representative academic developments from classical EF mechanisms and Fe²⁺ regeneration to advanced catalyst/electrode design, magnetic-field-assisted electrochemistry, and spin-chemistry regulation.

Most existing reviews focus either on specific materials or on magnetic fields in general, and do not give a clear picture on how magnetic fields act on the main reactants in the EF system. To understand the existing gaps, important recent publications in three areas are compared, as shown in **Table 1**: EF mechanisms, magnetic field effects in electrochemical systems, and spin chemistry phenomena in catalysis. A reference review on the fundamentals of EF mechanisms and Fe³⁺ regeneration was published by

some of us in 2023 ^[16]. It discusses electron transfer and mass transport strategies, including the effect of magnetic fields. However, it mainly attributes their benefits to physical improvements in diffusion, while paying little attention to possible spin-chemistry effects. Reviews on the use of magnetic fields in electrochemistry mostly focus on the MHD effect and related physical forces ^[11]. These reviews treat EF as a mere conventional electrochemical process, without considering its specific features, such as the complex interfaces and radical reactions involved. Regarding the recent reviews on spin chemistry in electrocatalysis, these have shown that electron spin states influence adsorption energies and reaction pathways ^[17, 18]; however, these works focus almost entirely on water splitting and fuel cell reactions. As far as we are concerned, these ideas have not been applied to EF systems for water treatment.

Table 1. Comparison of core dimensions and decoupling depth between existing authoritative reviews and this work.

Reviews in the Literature	Core Focus	Key Physicochemical Effects Involved	Specificity to EF Systems	Depth of Decoupling of Macroscopic Mass Transport and Microscopic Spin Chemistry
Fe ²⁺ regeneration ^[16]	Mechanisms of Fe ²⁺ regeneration	Electron transfer, macroscopic mass transport (including magnetic fields), photoelectrocatalytic coupling	High (Exclusively focused on EF systems)	Limited (No discussion on quantum spin effects)
Magnetic-field electrochemistry ^[20]	Universality of magnetic fields in electrochemistry	Lorentz force, Kelvin force, micro-MHD	Moderate (General electrodeposition and catalytic systems)	Moderate (Emphasizes macroscopic convection, but ignores intrinsic kinetic differences)
Spin-controlled electrocatalysis ^[21]	Electronic structure and spin chemistry	Orbital spin polarization, spin-selective electron transfer	Limited (Mainly targets water splitting/fuel cells involving OER/ORR)	High (Focuses on decoupling spin and non-spin effects in catalysis)
This review	Magnetically-enhanced EF	MHD, Kelvin force, micro-MHD, RPM, CISS	High (It considers Fe ²⁺ /Fe ³⁺ , H ⁺ , O ₂ , e ⁻)	High (Serves as the core logical thread running throughout the entire review)

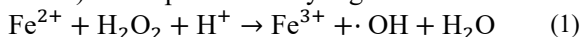
This review contributes in three important ways. First, it moves the field beyond the long-standing “black box” view of magnetic field effects by clearly separating macroscopic MHD mass transport from quantum spin-chemistry effects on intrinsic reaction kinetics. This decoupling directly addresses the central debate in the field and points the way to new experimental designs, such as using microelectrodes or parallel-flow cells to isolate spin effects from convection. Second, it replaces traditional material-based or electrode-based classifications with a unified four-species framework (Fe²⁺/Fe³⁺, H⁺, O₂, and e⁻). This species-by-species approach brings order to a complex gas-liquid-solid system and makes the analysis of coupled physical and quantum phenomena much clearer. Third, it introduces frontier spin chemistry concepts —such as the radical pair mechanism (RPM), chiral-induced spin selectivity (CISS), and control of intersystem crossing (ISC) — into environmental electrocatalysis for the first time.

This opens the door to selective generation of high-reactivity species such as singlet oxygen (¹O₂) or high-valent iron-oxo species (Fe(IV)=O) ^[19, 20]. The structure of the manuscript first covers the basic principles of EF process under magnetic fields; it then examines how magnetic fields influence the iron cycling, proton transport, oxygen activation, and electron transfer in turn; finally, these results serve to separate the contributions of MHD and spin chemistry and discuss future opportunities.

2. Mechanisms of magnetic-field influence on Fe²⁺/Fe³⁺ cycling

Considering the four-species framework established, we begin our species-by-species analysis with the Fe²⁺/Fe³⁺ pair and protons (H⁺). These species directly control the generation of •OH and the stability of the

entire catalytic cycle. Fenton's reaction requires sufficient Fe^{2+} and an acidic local environment (eq. 1), yet the electrode interface frequently suffers from Fe^{3+} accumulation and rapid H^+ depletion. External magnetic fields offer a powerful, non-contact means to enhance the mass transport of both types of species [11] while potentially modulating their intrinsic reaction kinetics through spin effects [22]. In the following sections, we examine these phenomena in detail, aiming to separate the contributions of MHD (i.e., convection) from spin-chemistry regulation.



2.1. Regulation of $\text{Fe}^{2+}/\text{Fe}^{3+}$ behavior via magnetic fields

2.1.1. Regulation of $\text{Fe}^{2+}/\text{Fe}^{3+}$ mass transport

Efficient cycling between Fe^{2+} and Fe^{3+} is essential in the EF process. The Fe^{3+} produced in the Fenton's reaction (1) must reach the cathodic sites to be reduced back to Fe^{2+} . This transport step often limits the overall reaction rate. External magnetic fields overcome this limitation by enhancing convection. Magnetic fields act on iron ions through three forces (Fig. 3).

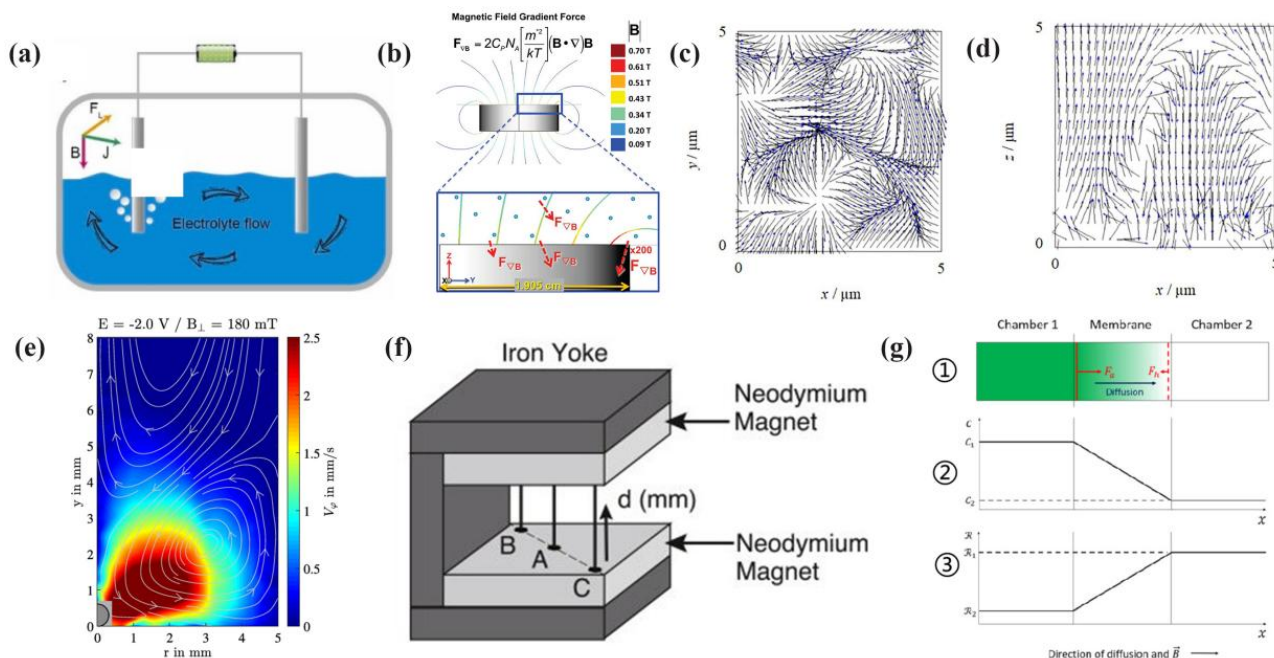


Fig. 3. (a) Scheme of electrolyte convection induced by the MHD effect. Modified with permission [24]. Copyright 2024, Elsevier. (b) Magnetic field gradient force acting on a fluid containing paramagnetic species with a uniform concentration. Modified with permission [25]. Copyright 2010, ACS. (c) Horizontal distribution of micro-MHD flow. Modified with permission [26]. Copyright 2019, Elsevier. (d) Cross-sectional distribution of micro-MHD flow. Modified with permission [26]. Copyright 2019, Elsevier. (e) Macroscopic mixing effect driven by MHD. Modified with permission [27]. Copyright 2016, ECS. (f) Design of a magnet device. Modified with permission [28]. Copyright 2024, Elsevier. (g) ① The diffusion process of an unstirred paramagnetic electrolyte from chamber 1 to chamber 2 through a thin, inert membrane under the influence of a homogeneous static magnetic field, B_0 . ② Illustration of the concentration profile across the membrane. ③ Illustration of the reluctance profile across the membrane. Modified with permission [29]. Copyright 2021, MDPI.

(1) Lorentz Force (MHD effect)

The Lorentz force drives convection in the electrolyte and directly accelerates the transport of Fe^{3+} to the cathode and Fe^{2+} back into the reaction zone [23]. In the solution bulk, it produces macroscopic mixing (Fig. 3a). Conversely, near the electrode surface or in the presence of gas bubbles, non-uniform current distributions create intense local micro-vortices (Fig. 3c, d), which is called micro-MHD effect. Unlike bulk MHD, which stirs the entire solution, micro-MHD

generates strong local mixing right at the three-phase interface. These micro-vortices are particularly effective at thinning the Nernst diffusion layer for Fe^{3+} and Fe^{2+} ions, greatly speeding up their delivery to and from the electrode surface [24].

The Lorentz force is determined by eq. 2:

$$F_L = q(E + v \times B) \quad (2)$$

Here, q is the charge of the ion, E is the electric field vector, v is the velocity vector of the ion, and B is the

magnetic flux density vector. The cross-product $\mathbf{v} \times \mathbf{B}$ produces a force perpendicular to both the ion motion and the magnetic field.

(2) Kelvin Force

Both Fe^{2+} (high-spin d^6) and Fe^{3+} (high-spin d^5) are paramagnetic. In a non-uniform magnetic field (**Fig. 3b**), they therefore experience a Kelvin force that attracts them towards regions of higher field strength. This force can induce mass transport and create concentration gradients. For example, Waskaas and Kharkats modeled mass transport driven by a magnetic force arising from gradients in iron ion concentration [29]. Similarly, studies on zero-valent iron (ZVI) corrosion showed that even weak magnetic fields promoted Fe^{2+} release, an effect attributed partly to the Kelvin force acting on the paramagnetic ions [30]. On magnetic catalysts, this creates steep local gradients, enriching iron ions directly at the active surface (**Fig. 3f**). The Kelvin force (F_K) is given by **eq. 3**:

$$F_K = (c\chi_m / 2\mu_0)\nabla B^2 \quad (3)$$

where μ_0 is the vacuum permeability, c is the molar concentration, χ_m is the molar magnetic susceptibility, and ∇B^2 represents the gradient of the square of the magnetic field strength.

(3) Paramagnetic Concentration-Gradient Force

Even in a uniform magnetic field, steep concentration gradients of paramagnetic species near the electrode (**Fig. 3g**) produce an additional force. This force drives iron ions toward regions that minimize magnetic reluctance and further enriches $\text{Fe}^{2+}/\text{Fe}^{3+}$ at the reaction zone. The force (F_c) is proportional to **eq. 4**:

$$F_c \propto B^2 \nabla c \quad (4)$$

where ∇c is the concentration gradient of the paramagnetic ions.

These three forces and their key characteristics in the EF systems are summarized in **Table 2**.

Table 2. Magnetic forces acting on $\text{Fe}^{2+}/\text{Fe}^{3+}$ in the EF systems.

Force Category	Governing Equation	Target Species	Characteristic Scale & Main Effect
Lorentz Force (Bulk & Micro-MHD)	$F_L = q(E + \mathbf{v} \times \mathbf{B})$	All moving ions	Macro: bulk convection Micro: micro-vortices at rough surfaces or via bubbles, ultra-thin Nernst layer
Kelvin Force	$F_K \propto \chi_m \nabla B^2$	Paramagnetic species (Fe^{2+} , Fe^{3+} , O_2)	Nanoscale interfacial enrichment on magnetic catalysts
Paramagnetic Concentration-Gradient Force	$F_c \propto B^2 \nabla c$	Regions with steep ∇c of paramagnetic ions	Mesoscale enrichment in the diffusion layer

The Lorentz and Kelvin force contribute differently to iron ion mass transport. The Lorentz force drives bulk convection that affects all charged particles and provides system-wide mixing (**Fig. 3e**). It is most effective at high current densities. In contrast, the Kelvin force is selective and acts primarily on paramagnetic iron ions. It can arise from external non-uniform magnetic fields (**Fig. 3f**) or from concentration gradients of the ions themselves, even in a uniform field (**Fig. 3g**). This force enriches or depletes ions at the electrode interface and is therefore most relevant for controlling the microenvironment near the surface. The two forces often interact: MHD-driven flow changes local concentration gradients, which in turn strengthens the Kelvin force. Careful reactor and magnet design can use this synergy to maximize iron ion transport.

Researchers have designed various magnetically enhanced electro-Fenton systems based on these forces, and representative examples of external magnetic-field regulation on iron species are summarized in Table S2 in the Supporting Information.

2.1.2. Alteration of $\text{Fe}^{2+}/\text{Fe}^{3+}$ catalytic activity

Although the magnetic forces above mainly improve

the physical mass transport of iron ions, magnetic fields can also directly change the intrinsic reaction kinetics of Fe^{2+} and Fe^{3+} . These kinetic effects directly control the generation efficiency of $\bullet\text{OH}$ (**Fig. 4a**). How to strictly separate true kinetic speed-up from the indirect effects caused by better mass transport remains a main challenge in this field.

Static magnetic fields speed up Fe^{2+} oxidation and H_2O_2 breakdown [10]. For example, weak magnetic fields removed arsenic faster and more completely than the control group (**Fig. 4b**). Hao et al. further showed that stronger magnetic fields promoted both Fe^{2+} to Fe^{3+} conversion and H_2O_2 breakdown at the same time [10]. Alternating magnetic fields give clear results as well [31]. Tafel analysis showed they lowered the catalyst Tafel slope from 97 mV dec⁻¹ to 62 mV dec⁻¹ (**Fig. 4c**). Wang et al. used a $\text{Fe}@\text{NiFe}_2\text{O}_4$ catalyst and found that alternating magnetic fields substantially increase bisphenol A degradation efficiency through magnetothermal effects and electric fields induced by nanoparticles. These apparent macroscopic kinetic accelerations mainly come from spin chemistry effects. Magnetic fields may modulate spin sublevel populations or spin-dependent pathways under specific magnetic/exchange-coupled environments. (**Fig. 4e**).

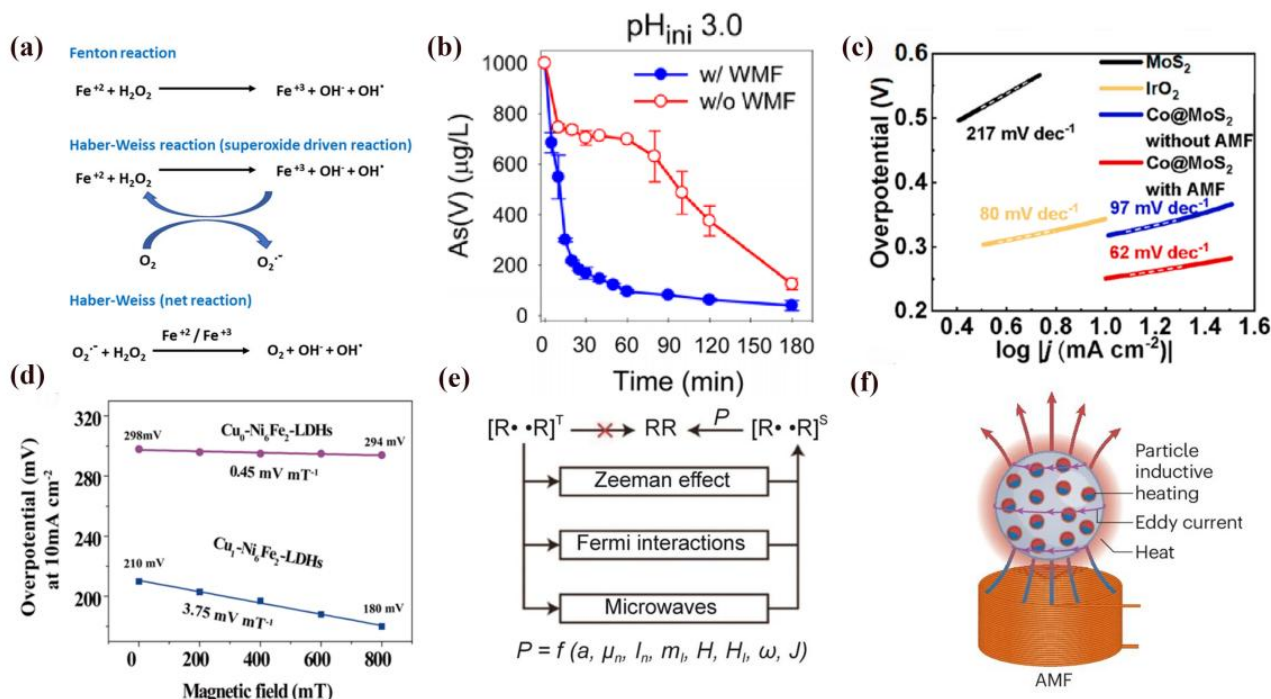


Fig. 4. (a) Basic free radical mechanisms of the Fenton and Haber-Weiss reactions. Modified with permission [39]. Copyright 2019, MDPI. (b) Influence of a weak magnetic field (WMF) on As(V) removal by ZVI. Modified with permission [30]. Copyright 2014, ACS. (c) Tafel plots of MoS₂, IrO₂, and Co@MoS₂ with and without an AMF. Modified with permission [40]. Copyright 2022, ACS. (d) Overpotential response of different LDHs catalysts under magnetic field. Modified with permission [32]. Copyright 2022, ACS. (e) General scheme of spin conversion induced by Zeeman and Fermi interactions and microwaves. The radical pair functions as a spin-selective microreactor in which the reaction probability p is a function of the magnetic parameters of the radicals (a, μ_n, I_n, m_b), magnetic fields (H, H_b, ω), and the exchange interaction J . Modified with permission [41]. Copyright 2024, Wiley. (f) The magnetothermal effect in liquid metal matrix composites (LMMCs) is induced by eddy currents in the liquid metal and inductive heating of magnetic particles under an alternating magnetic field (AMF). Modified with permission [42]. Copyright 2024, Springer Nature.

Sun et al. showed that by tuning the spin state of Fe (III), the catalyst response to magnetic fields became much stronger [32]. For example, Cu²⁺ doping shifts Fe (III) spin state to ferromagnetic and greatly raises the catalyst magnetic sensitivity (**Fig. 4d**).

In acidic EF systems, Fe³⁺ and Fe²⁺ are usually high-spin species because H₂O, OH⁻, and most oxygen-containing ligands provide weak ligand fields. Thus, Fe³⁺ can generally be regarded as a high-spin d⁵ center with $S = 5/2$, while Fe²⁺ is a high-spin d⁶ center with $S = 2$. Under ordinary static magnetic fields, the magnetic-field energy is too small to induce a universal low-spin/high-spin conversion of iron centers. Therefore, magnetic-field stabilization should be understood mainly as preferential population or alignment of Zeeman-split sublevels within an already accessible spin manifold, especially when the iron site is located in a magnetic or exchange-coupled environment [32].

This view helps reconcile conflicting reports on

Fe³⁺ reduction [11] When Fe³⁺ reduction is mass-transport limited, magnetic fields usually accelerate the apparent Fe³⁺ → Fe²⁺ regeneration by Lorentz-force-driven MHD convection, micro-MHD vortices, and Kelvin-force enrichment [23], which enhance Fe³⁺ delivery to the cathode and Fe²⁺ removal from the interface [29]. In this case, the positive magnetic effect is mainly transport-controlled. However, when Fe³⁺ reduction is kinetically controlled, the magnetic-field response may be affected by Fe(III) spin-state regulation and interfacial spin matching on magnetic or exchange-coupled surfaces [32]. Favorable spin-polarized electron injection from magnetic electrodes may lower the Fe³⁺ reduction barrier, whereas unfavorable magnetic-domain alignment, adsorption geometry, or exchange coupling may stabilize adsorbed Fe³⁺ or cause spin mismatch, thereby inhibiting electron transfer [32]. Therefore, magnetic fields do not universally accelerate or inhibit Fe³⁺ reduction; the observed

direction depends on whether transport enhancement, interfacial microenvironment changes, or intrinsic spin-dependent electron transfer dominates.

A key challenge is how to strictly separate these spin-driven kinetic effects from mass transport improvements [33]. Vensaus et al. (2024) built a special magneto electrochemical setup with non-magnetic electrodes [34]. By changing reactant concentration (high-concentration OER vs diffusion-limited ORR) and magnetic field strength in a controlled way, they separated mass transport contributions from intrinsic kinetic effects. They found that in diffusion-limited oxygen reduction (ORR), magnetic field enhancement exceeds 50% and comes mainly from Lorentz force-driven mass transport; in reactant-rich oxygen evolution (OER), the enhancement is small, showing limited intrinsic kinetic contribution. Xia et al. (2025) designed a thin-layer flow cell with high-speed forced convection [35]. On polycrystalline Au electrodes, electrolyte flows parallel to the surface at controlled high rates (up to 17.60 mL·min⁻¹). This fully suppresses natural diffusion and MHD convection to small, controlled changes. They isolated the real effect of magnetic fields on intrinsic overpotential and saw the pure kinetic drop in OER overpotential for magnetic electrocatalysts.

For alternating magnetic fields (AMF), both induced heating (magnetothermal effect) and non-thermal mechanisms must be considered. Wang et al. (2024) found in the Fe@NiFe₂O₄ Fenton-like system that AMF gave two contributions [36]: magnetothermal effect raises local temperature (~5-10 °C) and speeds up Fe(II)/Fe(III) conversion; the built-in electric field at nanoparticle interfaces (non-thermal) still promotes Fe(III) reduction even at fixed temperature and lowers the Tafel slope by ~35%. Trotsenko et al. (2026) latest review confirms that in AMF-enhanced electrocatalysis, induced heating (local heat from Néel/Brownian relaxation) works together with non-thermal spin polarization [37]. Even with strict temperature control ($\Delta T < 2$ °C), intrinsic activity still rises 20-30%, showing non-thermal mechanisms matter.

In addition, Fenton systems can form high-valent iron species such as Fe(IV)=O. Different reaction pathways may involve different spin states, suggesting that spin-state regulation of iron-based intermediates could potentially influence oxidant selectivity under specific catalytic environments [38].

Additional literature on magnetic-field regulation of iron ion activity is summarized in Table S3 in the Supporting Information.

3. Magnetic-field regulation of H⁺ behavior

Within the four core species framework, protons (H⁺) act both as the essential reactant that drives cathodic oxygen reduction to form H₂O₂ ($O_2 + 2H^+ + 2e^- \rightarrow H_2O_2$) and as the central microenvironment regulator that sustains both homogeneous and heterogeneous Fenton's reaction extent (eq. 1) [43]. At high current densities, however, rapid proton consumption at the cathode-electrolyte interface sharply lowers local H⁺ concentration, triggering pronounced interfacial alkalization. Such pH fluctuations not only suppress radical chain reactions but also induce Fe³⁺ hydrolysis and precipitation, ultimately causing electrode passivation and system failure [44].

Recent studies show that magnetic-field effects on proton behavior extend far beyond simple hydrodynamic stirring; they encompass a coupled progression spanning macroscopic continuum hydrodynamics, mesoscopic double-layer restructuring, and microscopic quantum-topological networks. To clarify the underlying mechanisms, the magnetohydrodynamic convective mass transport must be strictly decoupled from the field-induced modulation of intrinsic proton transport kinetics [45].

3.1. Macro- and micro-MHD effects on H⁺ mass transport

At the macroscopic scale, the most evident influence of the magnetic field on proton distribution arises from the MHD effect. When an external field (B) is applied to an electrolyte carrying a Faradaic current (j), moving ions experience a Lorentz force whose governing equation is eq. 2. This transverse body force induces convection in the bulk solution, accelerating H⁺ supply from the bulk to the electrode boundary layer and thereby mitigating diffusion-limited proton depletion. [45]. In the EF process, however, the dominant control of local interfacial pH occurs at the microscopic scale through the micro-MHD effect. Real catalyst surfaces contain roughness, pores, and evolving gas microbubbles (H₂ from hydrogen evolution or residual O₂). These heterogeneities distort the surface electric-field lines, producing highly non-uniform and multidirectional local current-density vectors. Interaction of these vectors with a uniform external field generates intense, localized micro-vortices near the gas-liquid-solid three-phase boundary [48].

Unlike bulk MHD, micro-MHD vortices function as countless nanoscale “stirrers” that act directly inside the Nernst diffusion layer. Their high kinetic-energy dissipation rapidly thins the diffusion layer to its limit and forcefully injects proton-rich fluid from the outer region onto active sites^[45]. Cyclic voltammetry and in-situ pH monitoring confirm that, under a parallel

magnetic field, interfacial proton consumption is rapidly compensated and the rise in surface pH is strongly suppressed (as shown in **Fig. 5a, b**). This forced micro-convection breaks the classical Fick-law diffusion limit, suppresses iron hydroxide precipitation in the local microenvironment, and thereby extends electrode lifetime and passivation resistance in EF operation.

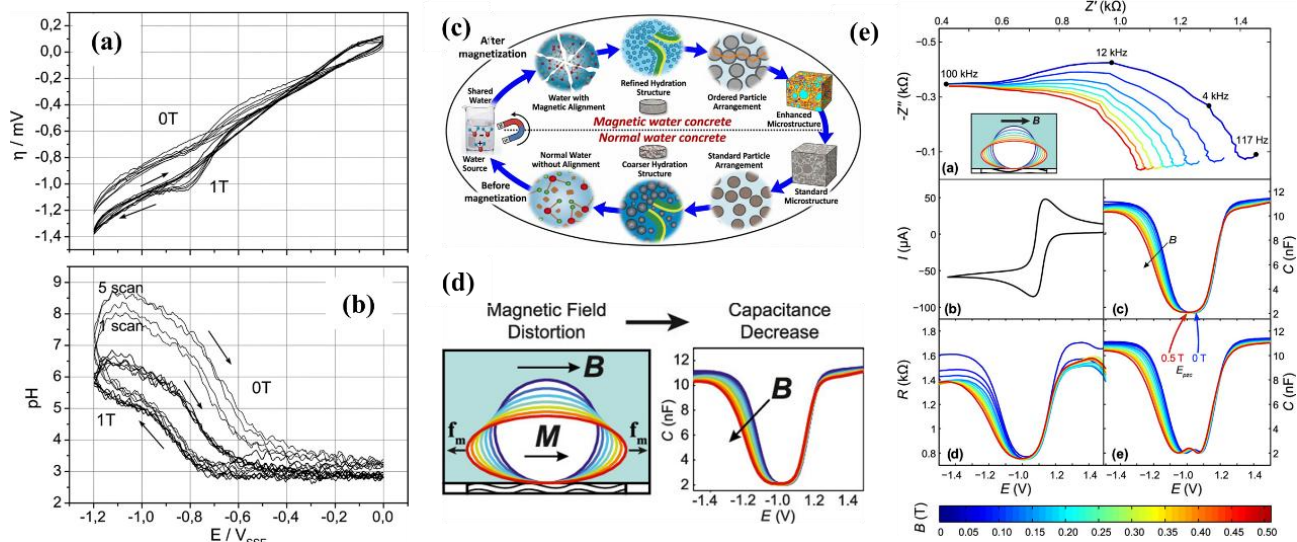


Fig. 5. Magnetic-field regulation of H^+ behavior. (a, b) Suppression of interfacial pH polarization and overpotential curves under applied field (0 T vs 1 T), revealing micro-MHD-driven forced proton injection and Nernst-layer compression. Modified with permission [45]. Copyright 2005, Elsevier. (c) Scheme of water-cluster restructuring before and after magnetization (large clusters $\rightarrow$ small clusters + hydrogen-bond rearrangement), demonstrating magnetic-field-induced depolymerization of the hydrogen-bond network. Modified with permission [46]. Copyright 2022, MDPI. (d, e) Double-layer capacitance C and charge-transfer resistance R versus potential at varying field strength (0 T vs 0.5 T), with explicit annotation of 0.25 nm OHP shift, Maxwell-stress force lines, and up to 50% change in C and R . Modified with permission [47]. Copyright 2019, ACS.

To decouple these hydrodynamic forces more clearly, **Table 3** classifies the magnetic-field force systems and their characteristic scales and effects on proton transport.

Table 3. Classification and characteristics of magnetic-field force affecting proton and related species transport in EF.

Force type	Governing equation	Target species	Characteristic scale	Core impact on proton transport and microenvironment	Reference
Bulk Lorentz force (Bulk MHD)	$F_L = j \times B$	All charged ions in bulk electrolyte	Macroscopic (cm)	Drives global convection; accelerates bulk H^+ replenishment to outer diffusion layer and reduces overall concentration polarization.	[23]
Micro-Lorentz force (Micro-MHD)	$F_{L, \text{micro}} = j_{\text{local}} \times B$	Non-uniform ion flow around bubbles/rough surface	Meso-/microscopic (μm)	Generates high-frequency local micro-vortices; sharply thins Nernst layer; violently disrupts gas-liquid-solid interfacial proton-depleted zones and suppresses local pH surge.	[48]
Kelvin force	$F_K = (\chi V / \mu_0)(B \cdot \nabla)B$	Paramagnetic species (O_2 , Fe^{3+} , O_2^-)	Nanoscale interface (nm)	Does not act directly on diamagnetic H^+ but enriches interfacial oxygen and iron ions, altering ORR and Fenton rates and indirectly modulating local proton consumption.	[49]

3.2. Kelvin-Force enrichment of paramagnetic species and indirect proton-consumption regulation

Although the Lorentz force directly drives H^+ transport, the Kelvin force provides indirect regulation

by gradient attraction of paramagnetic species. Its governing equation is $F_k = (\chi V / \mu_0)(B \cdot \nabla)B$. In the EF process, O_2 and Fe^{3+} are concentrated at the electrode surface while H^+ experiences mild repulsion. This

accelerates ORR (increasing proton consumption) and the $\text{Fe}^{2+}/\text{Fe}^{3+}$ cycle, forming a “faster consumption-stronger replenishment” balance with micro-MHD that suppresses interfacial alkalization and optimizes the proton-coupled electron-transfer pathway^[49].

3.3. Magnetic-field reshaping of water-cluster structure and hydrogen-bond network

After clarifying classical hydrodynamic transport, it is also useful to consider whether magnetic fields may influence the proton/solvent environment at the molecular level. Proton migration in aqueous solution depends strongly on intermolecular interactions. Liquid water forms a dynamic three-dimensional hydrogen-bond network, in which water molecules can assemble into clusters with different sizes and relaxation dynamics^[50].

Previous physicochemical and spectroscopic studies, including infrared, Raman, X-ray diffraction, and NMR analyses, have suggested that magnetic fields may perturb hydrogen-bond organization and water-cluster dynamics under certain conditions^[46, 50, 51]. These effects may involve partial rearrangement of water clusters, changes in rotational relaxation, and modification of the local solvation environment. As schematically shown in Fig. 5c, magnetic exposure has been proposed to induce partial restructuring of water clusters and hydrogen-bond networks^[52]. Such solvent-structure changes may provide a possible molecular-level background for discussing proton mobility in aqueous systems.

However, these observations should be interpreted with caution in the context of EF systems. Most available evidence for magnetic-field-induced water-cluster restructuring comes from bulk water or model aqueous systems rather than operating EF reactors. Therefore, such solvent-structure effects should not be regarded as direct evidence for enhanced EF performance. In magnetic-field-assisted EF, the most directly supported effect on proton availability remains MHD- or micro-MHD-driven H^+ replenishment near the electrode interface, whereas water-cluster restructuring provides only a possible molecular-level pathway that requires further verification.

From this perspective, magnetic-field-induced changes in hydrogen-bond networks may influence the local proton-transfer environment, but their quantitative contribution to EF reactions remains uncertain. This possible pathway must be separated from more direct effects such as bulk convection, micro-MHD mixing, bubble removal, local pH regulation, and redistribution of O_2 and Fe species near catalytic sites.

3.4. Potential magnetic-field regulation of Grotthuss proton transport

The core of decoupling the field's effect on intrinsic proton kinetics lies in the unique Grotthuss mechanism. Unlike most metal ions, such as Na^+ and Fe^{2+} , which migrate mainly through vehicular diffusion with their hydration shells, excess protons can be transported through structural diffusion along the hydrogen-bond network. In the Grotthuss process, excess protons do not travel long distances with any single water molecule; charge is relayed by rapid successive “hops” involving concerted cleavage and formation of covalent and hydrogen bonds. Microscopically, protons exist mainly in two limiting hydration structures: the Eigen cation (H_5O_4^+) and the Zundel cation (H_5O_2^+). Hopping consists of continuous interconversion between these states ($\text{Eigen} \rightarrow \text{Zundel} \rightarrow \text{Eigen}$).

Ab initio molecular dynamics and machine learning force field-simulations show that the rate-limiting step is not proton transport itself but reorientation and restructuring of the second hydration-shell hydrogen-bond network surrounding the defect^[53]. After a brief “burst” of multiple consecutive hops, the proton must wait for the next water molecule to rotate into an accepting orientation—the “resting period”^[54]. In principle, if a magnetic field changes the organization or rotational freedom of interfacial water molecules, it may indirectly influence the local proton-transfer environment. Although direct experimental evidence in electro-Fenton (EF) systems is still lacking, recent theoretical studies and molecular simulations suggest that magnetic-field-induced changes in the hydrogen-bond network may promote proton transport by facilitating water reorientation. Previous studies have suggested that magnetic fields may reduce the size of water clusters and alter the hydrogen-bond network, thereby potentially increasing the rotational freedom of hydration-shell water molecules^[54]. Previous studies have suggested that magnetic fields may alter water-cluster structure and hydrogen-bond dynamics^[52, 55-58], which provides a possible basis for discussing Grotthuss-related proton mobility. However, these effects have not yet been directly verified in EF systems. However, these effects have mainly been inferred from theoretical studies and indirect experimental observations, and have not yet been directly verified in electro-Fenton systems. When the magnetic field aligns with the electric field driving proton migration, the resulting charge-density rearrangement may promote more frequent formation of high-energy Zundel-like transition states, although this hypothesis remains to be experimentally validated.

Therefore, this mechanism should be treated as a possible pathway rather than as an established contribution to magnetic-field-assisted EF enhancement. Nevertheless, direct experimental evidence linking magnetic-field-induced water-structure changes to accelerated Grotthuss proton hopping in EF systems is still lacking. Future studies should combine operando local pH probes, isotope-

labeled proton-transfer experiments, ultrafast vibrational spectroscopy, NMR, neutron scattering, and molecular simulations to determine whether magnetic-field-induced hydrogen-bond restructuring contributes measurably to proton transport and EF performance. To highlight the differential impact of the magnetic field on these two mechanisms, **Table 4** summarizes their key features and field responses.

Table 4. Proton transport mechanisms in aqueous media and their responses to magnetic field-modulation.

Transport Mechanism	Physical/Chemical Nature	Rate-Limiting Step	Magnetic Field-Modulation and Response	Contribution in EF Systems	Reference
Vehicular Mechanism	Hydrated proton (e.g., HX, OX^+) migrates as a whole with its hydration shell via Brownian diffusion	Macroscopic solvent viscosity and Stokes drag	Field reduces cluster size, mildly lowers local viscosity, modestly increasing diffusion coefficient.	Minor; provides baseline transport only in high-electrolyte or moderate-viscosity regions.	[52]
Grotthuss Mechanism	Charge relay via covalent-bond cleavage and formation along the hydrogen-bond network	Second-hydration-shell hydrogen-bond rupture and water reorientation	Field-induced changes in water-cluster structure may alter rotational dynamics and the local proton-transfer environment, but direct EF evidence remains lacking.	Potentially important at the interface, but its contribution in EF systems remains hypothetical and must be separated from MHD-driven H^+ replenishment.	[54]

3.5. Emerging interfacial and proton-environment effects: evidence gaps and perspectives

Beyond MHD-driven proton replenishment, several interfacial and solvent-structure effects have been proposed to explain magnetic-field-assisted EF performance, but their direct evidence in EF systems remains limited. Therefore, these mechanisms should be regarded as emerging perspectives rather than established conclusions. First, Maxwell-stress-induced restructuring of the electrochemical double layer may alter interfacial capacitance [34], wettability [45], and the distance for proton-coupled electron transfer [47]. However, most evidence for this effect comes from general magnetochemical systems rather than EF reactors, and its contribution must be separated from MHD convection, bubble removal, local pH changes, and reactant enrichment [55]. Second, magnetic-field-induced changes in water clusters and hydrogen-bond networks may influence proton mobility and the Grotthuss mechanism [50], but the direct connection between such solvent-structure changes and EF reaction rates remains insufficiently verified [54]. Strong-field effects such as Paschen-Back-type perturbations should therefore be mentioned only as speculative possibilities under special field conditions, rather than as dominant mechanisms under typical EF operation [44]. Third, magnetic fields may help widen the apparent pH operating window by suppressing interfacial alkalization, promoting local proton replenishment, and maintaining soluble iron species [56]. It should be clarified that this does not mean that the magnetic field acidifies the entire bulk solution [45]. The

bulk pH may remain near neutral, whereas a transient acidic microenvironment can form within the thin electrode diffusion layer because H^+ , O_2 , and Fe species are redistributed near catalytic sites by MHD/micro-MHD convection and magnetic-gradient effects [57]. Therefore, the magnetic-field effect mainly regulates the local interfacial pH and species distribution rather than the average pH of the whole solution [44]. Nevertheless, pH-window expansion in EF is more directly supported by local microenvironment regulation [56], electrode design [57], and iron speciation [58] control than by Maxwell stress or quantum-level proton effects alone. Future studies should verify these emerging mechanisms using operando local pH probes, electrochemical impedance spectroscopy, differential capacitance measurements, isotope-labeled proton-transfer experiments, NMR/Raman analysis of water structure, and paired magnetic-field controls that minimize MHD effects. This more cautious interpretation avoids over-attributing EF enhancement to weakly established interfacial mechanisms.

These multi-scale mechanisms are supported by experimental and computational studies, and representative literature on magnetic-field effects on proton behavior is summarized in Table S4 in the Supporting Information.

4. Modulation of electron availability by magnetic fields

Following the analysis of magnetic regulation on the Fe^{2+}/Fe^{3+} cycle and H^+ transport, this section focuses

on electrons (e^-)—the intrinsic carrier driving all reactions in the EF process. As the link between macroscopic hydrodynamics and microscopic quantum spin, the electron transfer rate dictates cathodic O_2 reduction, Fe^{3+} regeneration, and overall system efficiency. A core scientific question remains: does the magnetic field affect electron behavior indirectly by enhancing reactant mass transport, or directly by altering the intrinsic electron transfer rate via spin effects? [17] Within the four-species framework, this section first isolates macroscopic hydrodynamic regulation driven by Lorentz, micro-MHD, and Kelvin forces (Section 4.1). It then examines quantum mechanisms, including spin polarization, the radical pair mechanism (RPM), CISS effect, and high-valent iron-oxo spin reconstruction (Section 4.2). This establishes a strict decoupling of macroscopic and microscopic effects.

4.1. Regulation of electron transfer

Applying a magnetic field to the EF process typically alters key electrochemical parameters. The limiting current density usually increases (Fig. 6a), while the Tafel slope, exchange current density, and charge transfer resistance may also change (Fig. 6b-d).

These variations often lead to reduced reaction overpotentials. However, as van der Heijden et al. [55] pointed out, in their alkaline OER study, that non-kinetic factors (e.g., bubbles, ohmic resistance, and ion gradients) can confound the observed Tafel slope, especially at high current densities. Although Fig. 6e visualizes OH^- redistribution rather than electron transfer itself, it is included here to illustrate how Lorentz-force-driven ion transport can indirectly regulate interfacial electron-transfer kinetics. By suppressing concentration polarization and compressing the diffusion layer, magnetic-field-induced convection changes the local availability of electroactive species and the interfacial electric field. These changes can decrease the apparent charge-transfer resistance (R_{ct}) and sustain Faradaic electron transfer at the electrode/electrolyte interface [38]. The resulting increase in the interfacial concentration of electroactive species facilitates interfacial Faradaic reactions, decreases the charge-transfer resistance (R_{ct}), and thereby sustains efficient electron transfer across the electrode/electrolyte interface. Therefore, the enhancement of ion transport serves as the physical basis for the observed improvement in electron-transfer kinetics rather than representing an independent transport phenomenon.

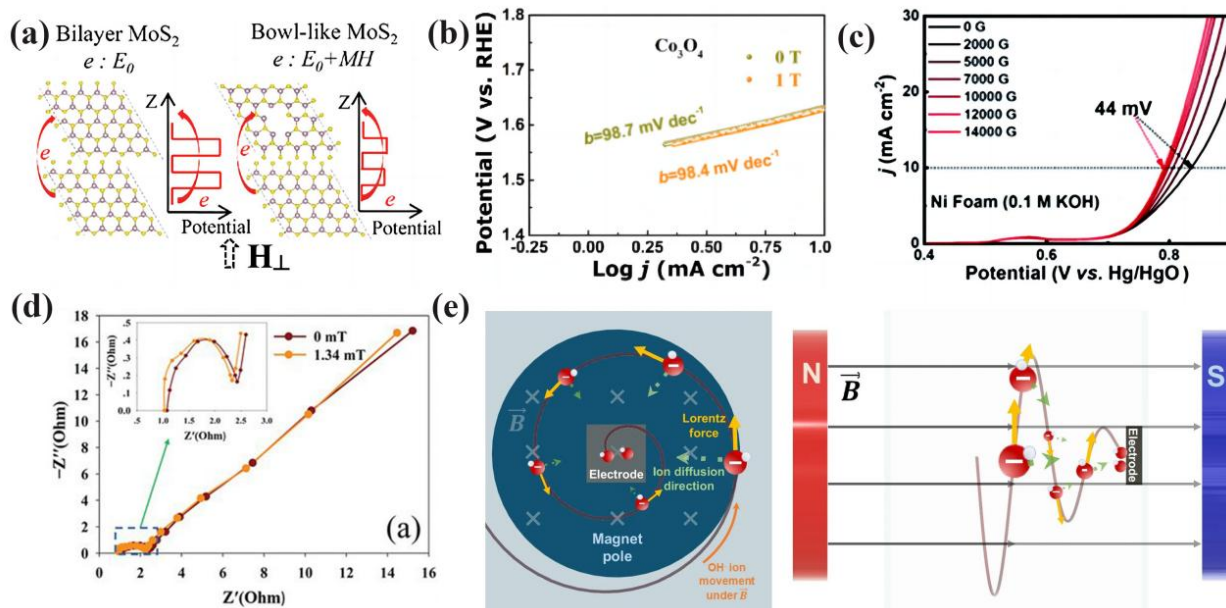


Fig. 6. (a) Electron transfer under a magnetic field for bilayer MoS₂ and bowl-like MoS₂. Modified with permission [59]. Copyright 2023, Wiley. (b) Tafel slopes of FeCo₂O₄ without and with a magnetic field. Modified with permission [60]. Copyright 2023, Elsevier. (c) Polarization curve of nickel foam under a magnetic field. Modified with permission [61]. Copyright 2022, Royal Society of Chemistry. (d) Nyquist plot of a MnO₂/electrospun carbon nanofiber electrode at 1.34 T and 0 mT. Modified with permission [62]. Copyright 2017, Elsevier. (e) Schematic illustration of Lorentz-force-driven OH^- redistribution near the working electrode (WE), used here as an example of how magnetic-field-induced ion transport can influence interfacial concentration polarization and apparent electron-transfer kinetics. Modified with permission [34]. Copyright 2024, Springer Nature.

4.1.1. Lorentz force-driven macroscopic and microscopic convection (bulk and micro-MHD)

The most direct physical effect of an external static magnetic field on moving charges (including electrolyte ions and migrating electrons) is the Lorentz force. This body force, perpendicular to both current and magnetic fields, induces global convection in the bulk electrolyte (Navier-Stokes equations). Macro-MHD convection disrupts the concentration polarization layer, accelerating the transport of dissolved oxygen and iron ions from the bulk solution to the electrode. At the electrode surface, this macroscopic convection compresses the Nernst diffusion layer^[20]. The reactant concentration gradient increases the limiting diffusion current density. This indirectly manifests as increased electron throughput in the system.

However, microscale hydrodynamic imbalance (micro-MHD) often determines electron transfer efficiency during actual EF polarization. Real electrode surfaces (e.g., porous carbon felt or nanoparticle substrates) are geometrically uneven and continuously generate microbubbles. These bubbles cause the local current density vector (j_{local}) to become highly multidirectional and non-uniform. The interaction between these distorted local currents and the uniform external magnetic field generates high-frequency, dissipative micro-vortices at the gas-liquid-solid interface^[20]. The microscopic Lorentz effect acts as a local "vortex pump" near active sites, rapidly expelling ions (e.g., OH^-) near the outer Helmholtz plane (OHP). This microscopic convection breaks the natural diffusion limits set by Fick's law. It minimizes concentration polarization during charge transfer, enabling continuous, high-speed electron transfer that would otherwise stall under standard reactant-depleted conditions^[34].

4.1.2. Bubble dynamics evolution and dynamic maintenance of electroactive surface area

At high current densities, bubble-evolving side reactions (e.g., hydrogen evolution) occur on the electrodes of EF systems^[63]. Bubble nucleation, growth, and detachment physically negatively affect the electron transfer. Because bubbles are highly insulating, a minor increase in coverage masks catalytic active sites^[64]. This reduces the electrochemically active surface area (ECSA) and increases local ohmic resistance, suppressing electron transfer^[65]. Magnetic fields alter bubble dynamics through hydrodynamic effects^[66]. Magneto-electrochemical studies using high-speed cameras and in situ visualization show that local

fluid vortices induced by macro- and micro-MHD apply strong hydrodynamic shear forces to attached bubbles. This shear force disrupts the force balance between surface tension and buoyancy at the solid-liquid interface, forcing bubbles to detach early. Using a high-speed flow cell, Vensaus et al.^[34] observed that accelerated bubble detachment under a magnetic field macroscopically expands the dynamic electrode active area. The decreased bubble residence time continuously renews the catalytic interface. This stabilizes the gas-liquid-solid three-phase boundary, maintaining efficient electron transfer^[67].

4.1.3. Kelvin force and interfacial gradient enrichment of paramagnetic species

As explained above, beyond the Lorentz force acting on moving charges, external magnetic fields and their gradients directly affect paramagnetic molecules via the Kelvin force^[68]. In the EF systems, the core O_2 precursor (a ground-state triplet) and iron intermediates (high-spin Fe^{2+} and Fe^{3+}) are highly paramagnetic. For ferromagnetic electrode materials (e.g., Fe_3O_4 or CoFe alloy nanoparticles), an external magnetic field aligns internal magnetic domains, generating steep local magnetic field gradients at the catalyst-electrolyte nanoscale interface. This microscopic gradient induces a directional Kelvin force that concentrates paramagnetic species (O_2 and Fe^{3+}) from the bulk solution to the electrode surface^[11]. The local interfacial concentration of paramagnetic reactants can exceed their bulk saturation limit. According to the Butler-Volmer equation, this elevated interfacial reactant concentration directly increases the local exchange current density and decreases the charge transfer resistance (R_{ct}). Therefore, the Kelvin force alters the thermodynamic distribution of paramagnetic species, indirectly accelerating electron consumption and transfer^[49].

4.1.4. Magnetothermal effects and induced electric field driving of alternating magnetic fields (AMFs)

Compared to static magnetic fields, alternating magnetic fields (AMFs) introduce an additional mechanism for electron regulation^[37]. For catalysts containing ferromagnetic nanoparticles or single-atom magnetic sites, AMFs induce a magnetothermal effect^[40]. Trotsenko et al.^[37] noted that the internal magnetic moments of ferromagnetic nanoparticles continuously flip under high-frequency AMFs. Energy dissipation via Néel and Brownian relaxation converts electromagnetic energy into localized heat. This internal heating rapidly raises the microenvironment

temperature at the catalyst surface (local $\Delta T < 10\text{ }^{\circ}\text{C}$), while the bulk solution temperature remains unchanged. Following the Arrhenius equation, this elevated interfacial temperature accelerates the electron transfer rate and the $\text{Fe}^{2+}/\text{Fe}^{3+}$ redox kinetics. Additionally, AMFs induce non-thermal magnetoelectric effects [69]. High-frequency AMFs generate Faraday eddy currents and induced micro-electric fields within conductive carbon supports or metal catalysts [37]. In single-atom Fe/rGO systems, for instance, AMF-induced charge separation produces low-energy holes (E_{HOMO}) and excited electrons (E_{LUMO}), driving magnetovoltic and magnetoelectric activity. This induced micro-electric field has been proposed to facilitate the iron valence cycle and H_2O_2 activation without an external bias, thereby providing a possible pathway for converting magnetic energy into electron-transfer potential [69]. However, because alternating magnetic fields (AMFs) can simultaneously induce magnetothermal heating and non-thermal spin-related effects, distinguishing their respective contributions remains a significant experimental challenge. Based on the current understanding of magnetic-field-assisted electrocatalysis, a temperature-matched control protocol may provide a practical strategy for isolating the non-thermal spin contribution. Specifically, both bulk and local interfacial temperatures should first be monitored in real time under AMFs. An isothermal control experiment can then be performed by externally heating the electrolyte to reproduce the same temperature increase in the absence of a magnetic field. Subsequently, the catalytic performance of magnetic catalysts should be compared with that of non-magnetic counterparts possessing similar morphology, conductivity, and electrochemically active surface area. Finally, operando spin-sensitive characterization techniques, such as electron paramagnetic resonance (EPR), magnetic circular dichroism (MCD), or X-ray magnetic circular dichroism (XMCD), may be combined with electrochemical measurements to directly probe spin-state evolution [70]. If the enhancement observed under AMFs exceeds that of the temperature-matched control while correlating with changes in spin-sensitive signals, the remaining enhancement could be reasonably attributed to non-thermal spin effects. Although such a protocol has not yet been systematically implemented in electro-Fenton systems, it represents an important research direction for clarifying the intrinsic mechanism of AMF-assisted electron transfer.

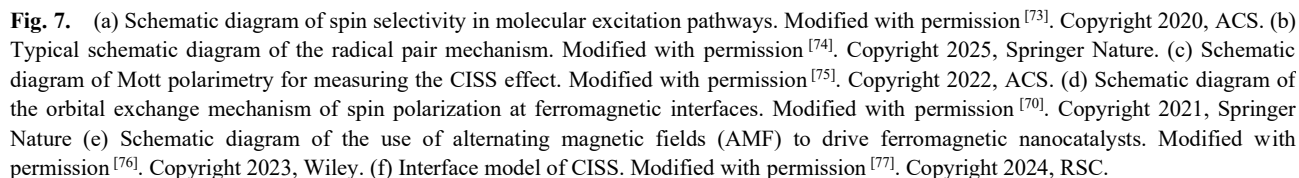
4.2. Regulation of electron activity and spin effects

Surpassing the classical limitations of mass

transport, tuning electron spin properties introduces a degree of freedom to drive chemical reactions [17]. By controlling spin states to alter electron transfer barriers and pathways, external magnetic fields directly improve magnetic-field-assisted electro-Fenton (magnetic-field-assisted EF) performance [70]. For instance, applying a magnetic field can activate otherwise "spin-forbidden" pathways, bypassing the intersystem crossing (ISC) energy penalty inherent to "spin-allowed" routes, thereby maximizing energy efficiency (**Fig. 7a**). Mechanistically, this enhancement proceeds via two primary pathways. First, spin-polarized electrons injected from ferromagnetic electrodes selectively couple with paramagnetic reactants (e.g., O_2) or intermediates, optimizing reaction kinetics and selectivity [17]. Second, within the radical pair mechanism (RPM), magnetic fields modulate the ISC rate between singlet (S) and triplet (T) states. This shifts radical pair lifetimes and subsequent product distributions (**Fig. 7b**) [71]. Additionally, the chiral-induced spin selectivity effect, where chiral structures act as electron spin filters, provides a related interfacial mechanism verifiable by Mott scattering (**Fig. 7c**) [72]. Many studies on spin effects, particularly in the field of catalysis, often rely on ferromagnetic catalysts. This raises a fundamental question: Does the magnetic field enhance the catalyst's inherent spin-related properties—such as spin-polarized surface states—associated with its ferromagnetism (as illustrated in **Fig. 7d**), or does the catalyst's ferromagnetism primarily act as an amplifier by concentrating magnetic flux lines to generate strong local magnetic field gradients or produce heat more efficiently under AMF conditions, as demonstrated in the experimental results shown in **Fig. 7e**. Recent studies indicate that the CISS effect can be regarded as a spin polarization effect induced by molecular configurations (e.g., electric dipole moment orientation) at interfaces, and this effect persists even in tunnel junctions without direct contact (see **Fig. 7f**). This provides physical modeling support for how transient local structures influence electron spin via analogous mechanisms.

4.2.1. Spin selectivity and breaking spin-forbidden barriers in ORR and OER

$2e^-$ ORR is the core in the EF process for reagents generation, yet its efficiency is fundamentally hindered by a significant "quantum energy barrier" [17]. Specifically, the reactants (H_2O , OH^-) and target product (H_2O_2) are diamagnetic singlets ($S = 0$), whereas ground-state oxygen ($^3\text{O}_2$) is a paramagnetic triplet ($S = 1$) [70]. According to the Wigner spin



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mismatch occurs [78]. Electrons encounter intense Pauli repulsion when entering specific π orbitals, resulting in a low transfer probability [79]. Overcoming this repulsion requires a spin-flip energy cost of approximately 1.1 eV, which is the quantum origin of high ORR/OER overpotentials [80].

Magnetic fields combined with ferromagnetic (FM) or ferrimagnetic (FiM) catalysts can effectively break this limit [17]. Under an external magnetic field, external fields align magnetic domains and rearrange d-orbital electrons near the Fermi level, inducing spin alignment [70]. These spin-polarized electron flows match the spin requirements of O_2 π orbitals, creating spin-selective electron transfer channels. This alignment directly bypasses the energy cost of spin-flipping. For example, experiments on Ag/Ni modified electrodes show that if the Ag layer is thinner than the spin diffusion length (~ 130 nm), polarized electrons cross the interface lossless, significantly enhancing H_2O_2 selectivity and activity [78].

4.2.2. Radical pair mechanism (RPM) and lifetime control of reactive oxygen species spin evolution

The EF system generates reactive oxygen species via the Fenton's reaction (1) [81]. This involves transient radical pairs (RPs), such as $[Fe(III)-OOH]$ complexes. The radical pair mechanism (RPM) explains how weak magnetic fields influence reaction rates at the sub-atomic scale [8]. Nascent RPs in the "solvent cage" maintain spin correlation as either singlets (S) or triplets (T). Without a magnetic field, singlets interconvert with the three triplet sub-levels ($T_{\{+1\}}$, T_0 , $T_{\{-1\}}$) via hyperfine coupling (HFC). Singlet RPs often undergo "back-electron transfer" or recombination into inactive ground-state molecules, leading to reagent waste. Applying an external magnetic field induces Zeeman splitting, separating the $T_{\{+1\}}$ and $T_{\{-1\}}$ energy levels [82]. This restricts the S state to mixing only with T_0 , significantly suppressing the intersystem crossing (ISC) rate. Consequently, RPs remain in the triplet state longer. Since triplets are spin-forbidden from direct recombination, the radicals gain a longer lifetime to undergo cage escape, becoming free $\bullet OH$ or $\bullet O_2^-$ [83]. RPM may contribute to ROS lifetime/selectivity, but direct evidence in EF remains limited for the same reagent consumption.

Although the RPM provides a plausible explanation for magnetic-field-enhanced ROS selectivity, direct detection of radical pairs under high-current EF conditions remains difficult [8]. Radical pairs are short-lived, spin-correlated intermediates confined within solvent cages or interfacial microenvironments, rather

than free ROS detected after cage escape. Their lifetimes are usually shorter than the time resolution of conventional steady-state spin-trapping EPR, and they may rapidly evolve into $\bullet OH$ [82], $O_2\bullet^-$ [83], $^1O_2$ [84], H_2O_2 [85], $Fe(IV)=O$ [86], or inactive recombination products [8]. High-current EF further introduces local pH gradients [45], Joule heating [55], bubbles [63], oxygen depletion [64], Fe precipitation [65], and electromagnetic noise [66], which can distort EPR signals and alter radical lifetimes. Therefore, RPM should be discussed as a plausible spin-chemistry pathway rather than a directly proven mechanism unless supported by spin-sensitive evidence and strict hydrodynamic control experiments [8, 34, 35, 71, 82, 83].

4.2.3. Spin state reconstruction of high-valent iron-oxo ($Fe(IV)=O$) and non-radical selectivity

Traditional EF systems rely on non-selective $\bullet OH$, which is easily quenched by background substances like Cl^- or HCO_3^- [84, 85]. To overcome this critical limitation, the development of non-radical oxidation pathways based on high-valent iron-oxo species ($Fe(IV)=O$ or $Fe(V)=O$) has emerged as a frontier strategy. $Fe(IV)=O$ displays two-state reactivity. Specifically, the high-spin ($S = 2$) or intermediate-spin ($S = 1$) states possess more unpaired d-electrons, thereby providing lower-energy electron transfer channels and markedly higher oxidation rates [86]. The spin-state regulation of high-valent iron-oxo species may provide a possible route for improving oxidant selectivity in EF-related systems. In principle, ligand-field engineering, defect regulation, or heteroatom coordination may tune the electronic structure and spin-state accessibility of $Fe(IV)=O$ species. Under magnetic or exchange-coupled catalytic environments, magnetic-field-related spin alignment may further influence electron-transfer pathways and oxidant selectivity. However, direct evidence for precise magnetic-field control of $Fe(IV)=O$ spin states in EF systems is still limited. Therefore, $Fe(IV)=O$ -related spin regulation should be regarded as a promising but still hypothesis-driven pathway rather than an established mechanism [86, 87].

4.2.4. The field-free revolution of chiral-induced spin selectivity

Generating magnetic fields via external electromagnets poses significant engineering challenges, such as high energy consumption and complex reactor design [19]. To address this limitation, chiral-induced spin selectivity (CISS) offers an alternative that enables spin-polarized electron injection without any physical magnets [88]. In the CISS

effect, electron transport through chiral molecules (e.g., DNA or chiral MOFs) induces strong intrinsic spin-orbit coupling. Consequently, one spin orientation (e.g., $\uparrow$) tunnels freely through the chiral potential, while the opposite spin experiences intense backscattering, creating highly efficient spin filtering at the molecular scale. When integrated into EF technology, CISS functions as a passive, built-in spin filter. By anchoring chiral molecules (e.g., BNP) as self-assembled monolayers (SAMs) on the cathode surface, the delivered current acquires 60%-90% spin polarization. These polarized electrons then perfectly match the triplet orbitals of paramagnetic oxygen, thereby

accelerating $2e^-$ -ORR toward H_2O_2 generation while thermodynamically suppressing spin-mismatched side reactions^[89]. Because CISS relies solely on molecular topology rather than external energy input, it provides a zero-energy, scalable pathway for distributed wastewater treatment.

To summarize the distinct mechanisms by which magnetic fields and CISS regulate electron spin polarization and transfer kinetics in EF systems, **Table 5** compares the representative pathways, including external field-assisted spin alignment, ferromagnetic/ferrimagnetic catalysts, and passive CISS-based spin filtering.

Table 5. Comparison of mechanisms by which magnetic fields influence electrons in electrochemical systems.

Mechanism	Key Theory / Principle	Primary Experimental Evidence	Potential Relevance and Impact on Key EF Steps	References
Electrode Spin Polarization	Consistent orientation of electron spins in a ferromagnetic material under a magnetic field	XMCD, spin-resolved photoemission spectroscopy confirming spin polarization on ferromagnetic surfaces.	If the cathode contains a ferromagnetic component, it could affect electron injection efficiency, altering the kinetics/selectivity of ORR or Fe^{3+} reduction.	[90]
Electron Spin Injection	Injecting spin-polarized electrons from a spin source (e.g., ferromagnetic electrode) into a non-magnetic layer or reactant.	Magnetoresistance effects in organic spintronic devices; C_{60} acquiring induced magnetic moment from Co.	Theoretically could affect electron transfer steps involving O_2 or Fe^{3+} , but the actual contribution is unknown.	[91]
Radical Pair Mechanism (RPM)	Magnetic field affects the singlet-triplet intersystem crossing rate of radical pairs, altering subsequent product yields.	Time-resolved EPR, CIDNP/CIDEP; magnetic field effects on product yields in photochemical/biochemical radical reactions.	If key radical pairs are formed, could affect the disproportionation/recombination of $\cdot OH$ or its reaction selectivity with other species.	[92]
Magnetic Field Effect on Catalyst Spin State	Magnetic field alters the d-orbital electron configuration and spin multiplicity of transition metal active centers.	Mössbauer spectroscopy, XAS/XES, magnetometry showing that magnetic fields or coordination environments can modulate the spin state of Fe in Fe-N-C.	Directly affects the adsorption energy and activation barrier of O_2 , H_2O_2 , etc., on the catalyst surface, thereby regulating the efficiency of ORR to H_2O_2 and the rate of H_2O_2 activation by Fe^{2+} .	[93]
Maxwell Stress	Magnetic field acts on paramagnetic species at the interface, creating stress that alters the double-layer structure	Electrochemical double-layer capacitance and charge transfer resistance change with the magnetic field	May alter the position of the Outer Helmholtz Plane (OHP), affecting electron transfer distance and rate.	[47]
Magnetic Field Gradient Effect	Magnetic field gradients at the electrode surface enhance electron transfer.	HER rate significantly increased on a diamagnetic electrode containing magnetic microparticles.	If the EF electrode generates or has a magnetic gradient, it could directly accelerate electron transfer steps.	[94]

4.3. Advanced in situ characterization and first-principles calculations for elucidating magnetic-field-assisted electro-Fenton mechanisms

Although considerable progress has been achieved in developing magnetic-field-assisted EF systems, the fundamental mechanisms governing catalytic enhancement under magnetic-field regulation remain incompletely understood^[38]. Unlike conventional electrocatalytic systems, magnetic-field-assisted EF involves the coupling of multiple physicochemical processes, including MHD convection, Fe^{2+}/Fe^{3+} redox cycling, oxygen reduction reaction, reactive oxygen species evolution, spin-dependent electron transfer, and interfacial mass transport^[12]. These processes dynamically evolve at the electrode/electrolyte

interface during electrolysis, making conventional ex situ characterization insufficient for capturing transient structural evolution and reaction intermediates. Therefore, advanced in situ/operando characterization techniques combined with first-principles calculations have become indispensable tools for directly correlating catalyst reconstruction, electronic structure evolution, interfacial reaction kinetics, and magnetic-field-induced catalytic enhancement, thereby providing fundamental insights into the working mechanisms of magnetic-field-assisted EF systems^[38].

4.3.1. In situ and operando characterization techniques

Advanced operando characterization techniques provide direct experimental evidence for understanding the dynamic evolution of catalysts and reaction

intermediates under magnetic-field regulation. Among them, in situ Raman spectroscopy has become one of the most widely employed techniques for monitoring catalyst reconstruction and structural evolution during electrochemical reactions [11]. Changes in Raman peak positions and intensities enable real-time identification of surface species, metal-oxygen bonding configurations, and intermediate products. In magnetic-field-assisted EF systems, operando Raman spectroscopy is particularly valuable for identifying magnetic-field-induced catalyst reconstruction, monitoring changes in metal-oxygen coordination environments, and correlating structural evolution with accelerated $\text{Fe}^{2+}/\text{Fe}^{3+}$ redox cycling and enhanced ROS generation.

Similarly, in situ Fourier-transform infrared (FTIR) spectroscopy provides molecular-level information regarding the adsorption and transformation of oxygen-containing intermediates and organic pollutants. Continuous monitoring of surface-bound species enables researchers to distinguish adsorption-controlled and reaction-controlled processes and to determine whether magnetic fields alter adsorption configurations, intermediate coverage, and reaction pathways through enhanced interfacial mass transfer and electron-transfer kinetics [34].

For iron-based electro-Fenton catalysts, in situ X-ray absorption spectroscopy (XAS), including XANES and EXAFS, has become one of the most powerful techniques for investigating the dynamic $\text{Fe}^{2+}/\text{Fe}^{3+}$ redox cycle. These techniques provide quantitative information on oxidation-state evolution, local coordination environments, and Fe-O bond distances during catalytic reactions. More importantly, operando XAS can directly verify whether external magnetic fields accelerate Fe^{2+} regeneration, stabilize catalytically active Fe sites, and facilitate continuous $\bullet\text{OH}$ production during long-term electrolysis.

Electron paramagnetic resonance (EPR/ESR) spectroscopy represents another indispensable technique for detecting short-lived reactive oxygen species [95]. Spin-trapping reagents such as DMPO and TEMP enable direct identification and quantification of hydroxyl radicals ($\bullet\text{OH}$), superoxide radicals ($\bullet\text{O}_2^-$), and singlet oxygen ($^1\text{O}_2$) [95]. Since magnetic fields influence electron spin polarization and radical-pair recombination, operando EPR provides one of the most convincing experimental approaches for validating spin chemistry mechanisms and distinguishing whether the enhanced catalytic performance originates from increased ROS generation or magnetic-field-induced spin regulation [71].

In addition, operando electrochemical techniques, including electrochemical impedance spectroscopy

(EIS), rotating ring-disk electrode (RRDE), differential electrochemical mass spectrometry (DEMS), and electrochemical quartz crystal microbalance (EQCM), provide complementary information regarding charge-transfer resistance, H_2O_2 generation efficiency, gaseous products, and interfacial mass variation [96]. In particular, operando EIS enables quantitative evaluation of the reduction in charge-transfer resistance resulting from magnetic-field-enhanced interfacial reactant concentration and accelerated electron transfer, whereas RRDE directly measures H_2O_2 selectivity. DEMS and EQCM further reveal gaseous-product evolution and dynamic catalyst reconstruction. The integration of these operando techniques provides comprehensive experimental evidence for understanding the synergistic effects of magnetic fields on electron transfer, mass transport, spin dynamics, and catalytic reactions [34]. These complementary probes and reactor-design considerations are summarized in **Fig. 8**, which highlights the distinction between material-focused and intermediate-focused measurements as well as reactor-design gaps and best practices for in situ/operando electrocatalysis [96].

4.3.2. First-principles calculations

Although advanced characterization techniques provide direct experimental observations, theoretical calculations, including DFT, spin-polarized DFT, and microkinetic modeling, have become useful tools for interpreting adsorption behavior, reaction energetics, and spin-dependent catalytic pathways in EF-related and oxygen-reduction systems [17]. DFT calculations can quantify the adsorption energies of O_2 , H_2O_2 , Fe species, and organic pollutants on catalyst surfaces, thereby identifying energetically favorable reaction pathways and clarifying how magnetic fields regulate interfacial catalytic processes [17].

Electronic-structure analyses, including density of states (DOS), projected density of states (PDOS), d-band center calculations, charge-density-difference mapping, and Bader charge analysis, provide valuable insights into electron redistribution induced by catalyst modification or magnetic-field regulation. These calculations establish quantitative relationships between electronic structures and adsorption behaviors, explaining experimentally observed improvements in electron-transfer kinetics and catalytic activity. More importantly, spin-polarized DFT calculations have recently emerged as a promising theoretical approach for revealing how magnetic fields regulate spin density, electronic configurations, adsorption energies, and reaction energetics, thereby providing theoretical support for magnetic-field-induced spin-selective

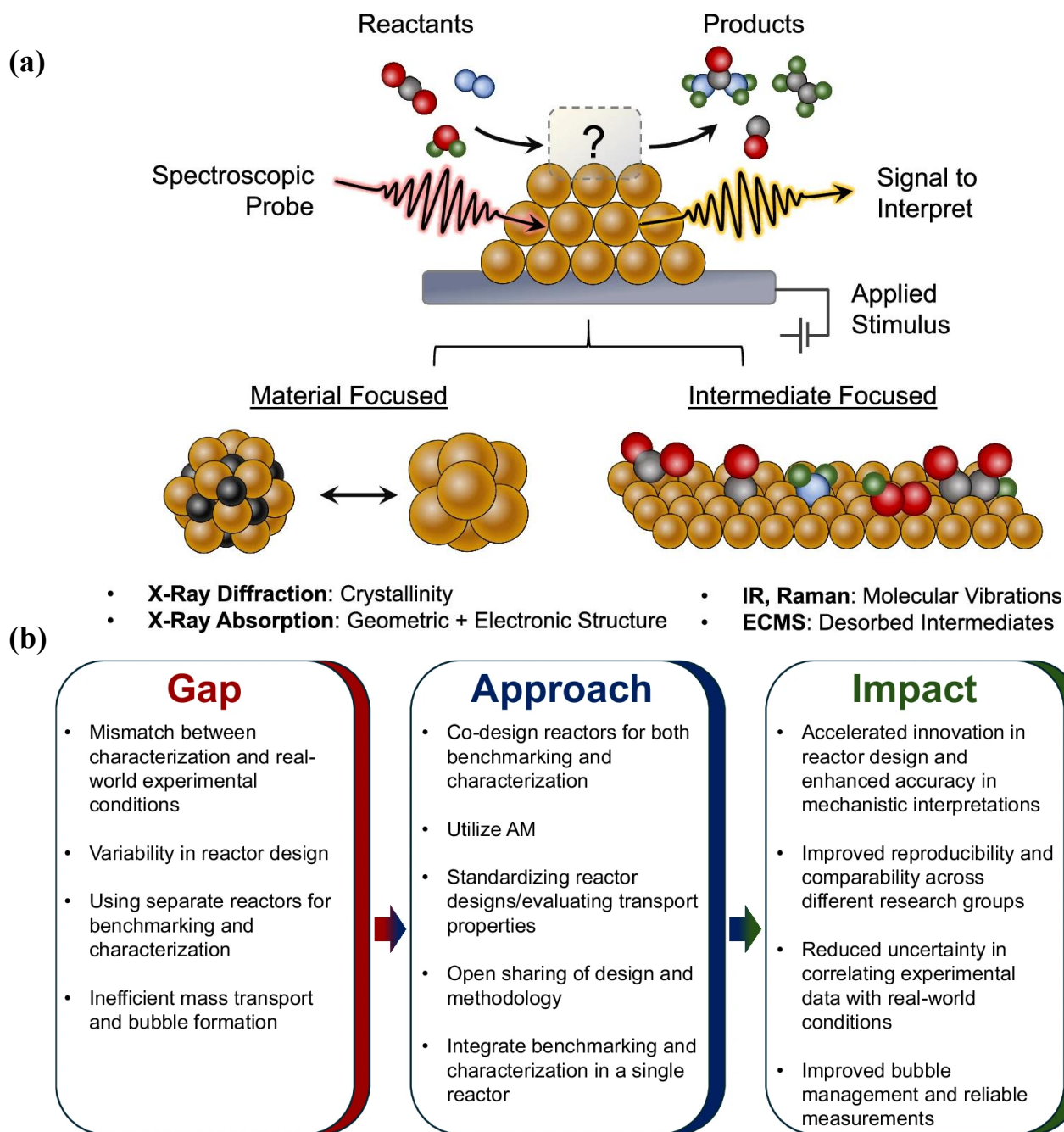


Fig. 8. Overview of in situ/operando analytical probes and reactor-design best practices for mechanistic interpretation in electrocatalytic systems. Reproduced from Prajapati et al. [96] under the Creative Commons Attribution-NonCommercial-NoDerivatives 4.0 International License.

catalysis [18]. The magnetic-field-altered eg-orbital occupancy reported by Jing et al. provides a representative example in which magnetic-field-induced electronic-structure regulation is linked to $2e^-$ ORR, H_2O_2 generation, and electro-Fenton water decontamination (Fig. 9) [6].

Reaction-energy calculations based on Gibbs free-energy diagrams can help identify rate-determining steps during oxygen reduction and Fenton-related reactions. In addition, climbing-image nudged elastic band (CI-NEB) calculations can be used to estimate activation barriers for elementary reaction steps [97].

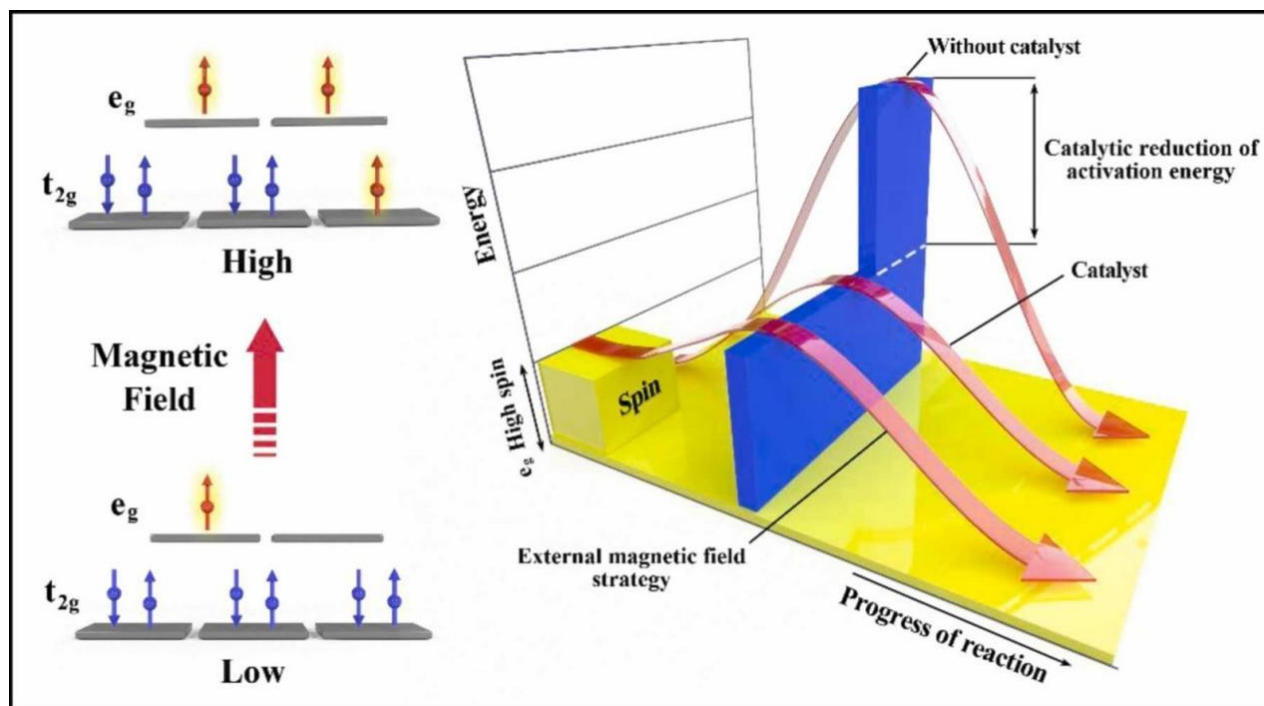


Fig. 9. Magnetic-field-altered eg-orbital occupancy for enhancing two-electron ORR and electro-Fenton water decontamination. Reproduced from Jing *et al.* [6] under the article's Creative Commons open-access license. Copyright 2025, The Authors. Published by Elsevier B.V.

Furthermore, ab initio molecular dynamics (AIMD) simulations have recently attracted increasing attention because they describe the dynamic evolution of catalyst/electrolyte interfaces under realistic reaction conditions [53]. AIMD simulations are particularly useful for investigating hydrogen-bond networks, proton transport, solvent reorganization, and magnetic-field-induced modifications of interfacial water structures. They can also visualize the dynamic transport of water molecules and Fe ions near electrode surfaces, thereby complementing experimental observations of MHD-enhanced mass transfer and interfacial regulation [34].

Together, these in situ/operando techniques and first-principles calculations provide complementary evidence for identifying magnetic-field-induced catalyst reconstruction, intermediate evolution, interfacial mass transport, and spin-dependent kinetics. A detailed summary of advanced operando characterization and theoretical calculation methods is provided in Table S5 in the Supporting Information.

5. Influence of magnetic fields on O₂

Following the analysis of the magnetic regulation

of the Fe²⁺/Fe³⁺ cycle, H⁺, and e⁻ injection, this section focuses on O₂, the most magnetically sensitive species in the EF process [7]. As the precursor for H₂O₂, O₂ faces a macroscopic mass transport bottleneck [98]. Its low solubility (~0.25 mM) causes rapid cathodic depletion, forcing the 2e⁻ ORR into a diffusion-controlled regime. Microscopically, O₂ also faces a quantum spin barrier. Ground-state O₂ is a spin-parallel triplet (³Σ_g⁻), while H₂O₂ and subsequent •OH usually involve singlet intermediates [99]. This triplet-to-singlet transition is strictly spin-forbidden [19]. Therefore, magnetic regulation of O₂ requires both macroscopic MHD mass transport and microscopic quantum spin control [11, 17]. To complete the four-species framework, Section 5.1 isolates physical mass transport driven by Lorentz, micro-MHD, and Kelvin forces. Section 5.2 then details spin chemistry pathways, including the radical pair mechanism, interfacial spin polarization, the CISS effect, and orbital hybridization.

5.1. Regulation of O₂ mass transport

Before assessing magnetic effects on intrinsic O₂ catalytic activity, one must separate the apparent current increases caused by mass transport [34]. In

standard EF processes, low O_2 solubility (~ 0.25 mM) causes rapid depletion at the cathode^[98]. This shifts the $2e^-$ ORR into a diffusion-controlled regime, limiting H_2O_2 yield and triggering side reactions like hydrogen

evolution^[100, 101]. External magnetic fields alter the hydrodynamic path of O_2 toward the gas-liquid-solid interface^[20]. This occurs via three physical forces: Lorentz, micro-MHD, and Kelvin force (**Fig. 10a**).

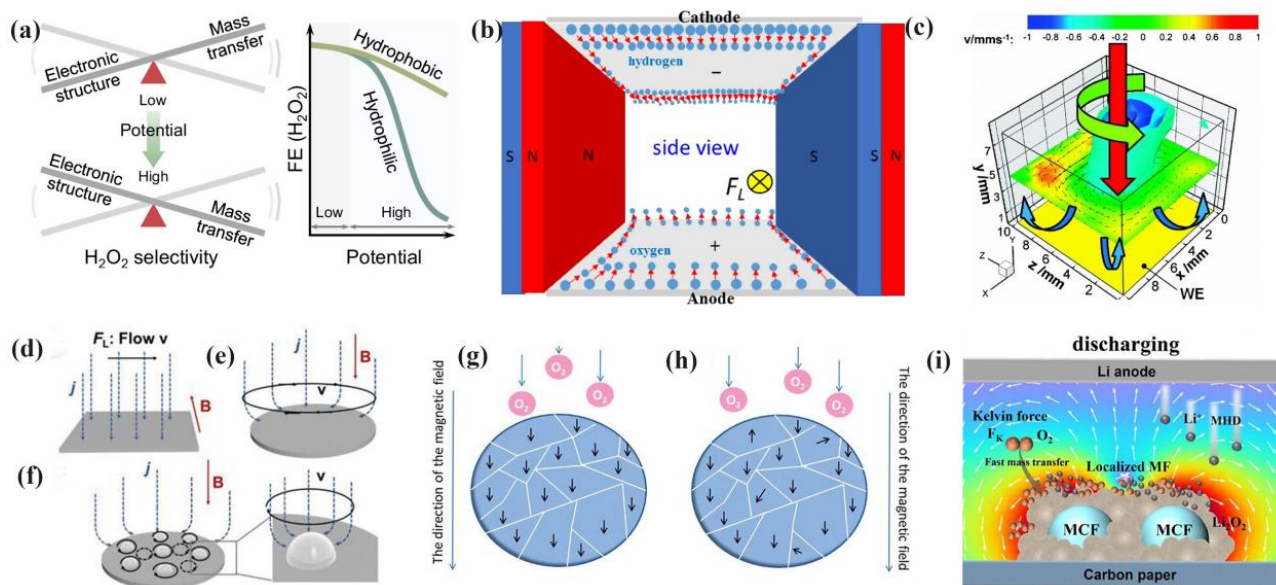


Fig. 10. Multiscale transport and oxygen-activation effects in magnetic-field-assisted EF systems. (a) Dominant factors controlling H_2O_2 selectivity at the electrode scale, adapted from Tang et al.^[112]. (b) Schematic representation of magnetic-field effects and Lorentz-force-driven MHD convection in electrochemical systems, adapted from Zhang et al.^[113]. (c) Micro-MHD phenomena occurring around a bubble when the magnetic field B is perpendicular to the current j , adapted from Tschulik et al.^[114]. (d) Suppressed Lorentz-force-driven convection under a parallel-field configuration ($B \parallel j$), which can be used as a control to minimize bulk MHD effects, and (e) MHD vortices forming around the electrode edge when B is perpendicular to j , adapted from Zhang et al.^[49]. (f) Micro-MHD phenomena around a bubble when B is perpendicular to j , adapted from Zhang et al.^[49]. (g, h) O_2 transport when an external magnetic field is applied to a magnetic catalyst, adapted from Wang et al.^[115]. (i) Discharge process of a magnetically enhanced Li- O_2 battery, illustrating the synergistic effects of MHD and Kelvin-force-driven oxygen transport, adapted from Chen et al.^[116].

5.1.1. Lorentz force-driven macroscopic convection and mass transport enhancement

When an external magnetic field is applied to an EF cathode carrying a Faradaic current, moving ions experience a Lorentz force ($F_L = j \times B$) perpendicular to the electric and magnetic fields^[34]. This body force ignores neutral O_2 . However, it drives bulk electrolyte rotation and macroscopic convection (macro-MHD)^[11, 20]. These pumps dissolved O_2 from the bulk solution to the hydrodynamic boundary layer (**Fig. 10b, 10d-f**). Recent magneto-electrochemical experiments quantify this enhancement via *in situ* visualization^[34]. In non-magnetic systems, Lorentz-induced ionic vortices dominate diffusion-limited reactions like the ORR. Studies show macro-MHD increases the mass transport-limited current density by 50% to 60% in ambient alkaline or acidic electrolytes. This directional pumping allows the ORR to access areas usually

inactive from O_2 depletion. In contrast, reactions with abundant reactants (e.g., OER) show minimal apparent gains under the same magnetic field due to negligible concentration polarization^[71]. This finding defines the boundary between mass transport and intrinsic activity in EF systems.

5.1.2. Microscopic stripping effect of Micro-MHD at the three-phase interface

Compared to bulk convection, micro-MHD provides localized stirring within the complex EF cathode microenvironment^[11, 102]. Even under a uniform external magnetic field, the interaction between distorted current vectors and the magnetic field generates high-frequency, dissipative micro- or nano-scale vortices at the gas-liquid-solid interface (**Fig. 10c**). Micro-MHD acts directly within the Nernst diffusion layer^[7]. Micro-vortices compress the O_2 diffusion layer to its physical limit. They also create a

severe local pressure differential across bubbles [103], accelerating the detachment of inactive bubbles. This rapid stripping reduces interfacial ohmic resistance and exposes fresh catalytic sites for O₂ adsorption. This amplifies the catalytic current without altering the intrinsic molecular energy barrier [7].

5.1.3. Nanoscale enrichment of paramagnetic oxygen dominated by Kelvin Force

While the Lorentz force stirs the bulk electrolyte, the Kelvin force acts as a targeted capture mechanism for O₂ [68]. Ground-state O₂ is paramagnetic, possessing a permanent magnetic moment and high molar magnetic susceptibility [104]. In a non-uniform magnetic field, O₂ experiences an attractive force toward maximum magnetic field gradients (**Fig. 10g, 10h**). Air is an ideal, low-cost oxygen source for gas-diffusion electrodes or open EF cathodes [105]. However, it contains abundant diamagnetic N₂ and water vapor [104-106], interfacial reactions often cause high concentrations of N₂ or liquid water to form a static layer on the electrode. This blocks continuous O₂ diffusion [107]. Introducing a non-uniform magnetic field or ferromagnetic catalysts (e.g., Ni foam, CoPt alloys, Fe₃O₄ nanocomposites) resolves this [108, 109]. External magnetic fields magnetize these materials, creating intense local gradients at rough surfaces, edges, or nano-tips [110]. Under these extreme gradients, the Kelvin force selectively pulls paramagnetic O₂ to catalytic sites. Thermodynamic repulsion simultaneously pushes diamagnetic N₂ and H₂O away

[104, 106]. This "magnetic molecular sieve" creates a local "O₂-rich microenvironment" on the catalyst without adding mechanical energy (**Fig. 10i**). *In situ* tracking confirms the physical synergy. Macro- and micro-MHD rapidly transport O₂ from the bulk to the near-interface. The Kelvin force then boosts the nanoscale concentration and anchoring of O₂ at catalytic sites [68].

Understanding these three forces presents a rigorous academic challenge: physical mass transport improvements are easily mistaken for enhanced intrinsic catalyst activity [11, 34]. In many reported non-decoupled systems, magnetic fields are linked to notable decreases in the ORR Tafel slope, thus shifting the onset potential positively [7, 110]. However, these apparent kinetic changes occur because MHD alters local ion concentration gradients, masking the true rate-determining step. To investigate spin chemistry impacts on intrinsic electron transfer rates, future EF research must employ decoupled magnetoelectrochemical setups [111]. Examples include using a high-speed rotating ring-disk electrode to impose hydrodynamic convection [111], overwhelming natural diffusion and MHD-induced convection. Alternatively, microelectrodes eliminate macroscopic fluid vortices. Special parallel magnetic and electric field configurations can even nullify the cross-product term [34]. Performance gains observed only under these controlled conditions, free from hydrodynamic interference, prove quantum-level magnetic regulation of intrinsic O₂ activity [34]. Table 6 summarizes these decoupling methods.

Table 6. Physical forces and decoupling features for O₂ mass transport at the EF cathode.

Force Type	Governing Equation	Target Species	Spatial Scale & Hydrodynamic Effect	Mass Transport Contribution & Decoupling Method
Macroscopic Lorentz force (Macro-MHD)	$F_L = j \times B$	All charged ions	Macroscopic (mm-cm): Large-scale vortices and directional flow	Breaks static diffusion limits; decoupled by RRDE forced convection
Microscopic Lorentz force (Micro-MHD)	$F_{L, \text{micro}} = j_{\text{local}} \times B$	Local ion flux at rough interfaces or bubbles	Mesoscopic (μm -nm): Micro-vortices at three-phase interfaces	Compresses diffusion layers and strips bubbles; decoupled via microelectrodes or parallel-field controls ($B \parallel j$) that minimize Lorentz-force-driven bulk convection
Kelvin force	$F_K = (\chi_m / \mu_0) c \nabla B^2$	Paramagnetic O ₂ and O ₂ ^{•−}	Nanoscale: Local magnetic gradients on catalysts	Selectively enriches O ₂ and repels N ₂ /H ₂ O; decoupled by comparing magnetic and non-magnetic substrates

5.2. Quantum control of intrinsic O₂ activation

After isolating MHD and Kelvin-driven mass transport using physical methods (e.g., RRDE forced convection, microelectrodes, or parallel fields), the research frontier shifts to quantum mechanics: Spin Chemistry [11, 34]. The ultimate goal of the EF cathode is the highly selective 2e[−] reduction of O₂ to H₂O₂ [98, 117, 118]. However, quantum physics poses a spin barrier [99]. This spin-forbidden process rarely

occurs spontaneously under standard thermodynamic conditions [119]. This causes the severe kinetic sluggishness and high overpotentials of O₂ activation in conventional electrocatalysis [17]. External magnetic fields alter deep physicochemical processes, including electron spin-orbit coupling, radical pair intersystem crossing (ISC), and interfacial quantum spin exchange [120, 121]. This directly reconstructs the kinetic reaction coordinates of O₂ reduction (**Fig. 10a**).

5.2.1. Quantum pathways of the radical pair mechanism and intersystem crossing

H₂O₂ generation at the EF cathode involves sequential proton-coupled electron transfer (PCET) steps [117, 122]. After capturing the first electron to form a superoxide radical (O₂^{•−}), it forms a spatially correlated "radical pair" (RP) with other interfacial paramagnetic species, such as a single transition metal electron at the catalyst center or a free hydrogen radical (H[•]) [17, 123]. The radical pair mechanism (RPM) is the core theoretical framework explaining how magnetic fields regulate O₂ reduction selectivity [71]. This magnetic effect is most pronounced at low O₂ concentrations where the radical pair lifetime is sufficient for spin evolution to occur (**Fig. 11b**); at high O₂ concentrations, the effect diminishes. These short-lived RPs exist in a coherent quantum superposition of a singlet state (S) and three triplet states (T₀, T⁺, T[−]) (**Fig. 11c**). Following the Pauli exclusion principle, the two unpaired electrons in the RP must have an antiparallel spin distribution [121, 124]. Only then can they enter the same bonding orbital to form singlet H₂O₂ or key singlet adsorbates like OOH [89]. Conversely, if the RP is in a parallel-spin triplet state, direct recombination to H₂O₂ is strictly spin-forbidden. The reaction is forced toward O-O bond dissociation pathways requiring higher activation energy (4e[−] reduction to H₂O) or side reactions yielding uncontrollable reactive oxygen species [71, 121].

External magnetic fields dictate the ISC rate between singlet and triplet RPs across two distinct dimensions: First, in the weak magnetic field regime, the hyperfine coupling (HFC) mechanism dominates [124]. Electron spins couple strongly with surrounding magnetic nuclei, driving mixing between the S and the three T states. Applying a specific external magnetic field lifts triplet degeneracy via the Zeeman effect [125]. This shifts the T⁺ and T[−] energy levels away from the S state, suppressing the mixing process [126]. This alters the statistical yield of RPs evolving into singlet or triplet states. Second, in the strong magnetic field regime, the Δg mechanism dictates spin dynamics [127]. The two distinct radicals in the RP (e.g., O₂^{•−} and a central metal site) possess slightly different g-factors. Under the same magnetic field, their Larmor precession frequencies differ [125–127]. This deviation causes a relative phase rearrangement of initially parallel electron spins, accelerating the triplet-to-singlet (T→S) conversion through non-thermodynamic means.

Applying specific static or alternating magnetic fields at the EF cathode accelerates T→S ISC, artificially accumulating singlet RPs [124]. Experiments

confirm that this RPM-based effect lowers the kinetic barrier for recombination and increases overall reaction current [7, 99]. Most importantly, it locks in ultra-high selectivity for H₂O₂ electrosynthesis [13], as demonstrated by recent RRDE tests. In oxide catalyst systems with paramagnetic centers (e.g., specific CeO₂ morphologies or CeMnCo composites), magnetic-induced spin evolution surges the 2e[−] ORR peroxide selectivity from ~50% at zero-field to over 90% [7, 13].

5.2.2. Catalyst interfacial spin polarization and quantum spin exchange interactions (QSEI)

While liquid-phase radical evolution addresses reaction intermediates, magnetic spin polarization of the solid cathode catalyst provides another pathway to bypass the O₂ activation barrier [70, 119]. To avoid the spin-flip energy barrier during O₂ reduction, the most direct strategy is providing a spin-selective charge transfer channel [79, 128]. This guides spin-matched electrons into the catalytic interface (**Fig. 11d**). When using ferromagnetic or ferrimagnetic materials (e.g., CoFe₂O₄, NiFe₂O₄, or N-doped carbon-encapsulated CoNi alloys with long-range magnetic order) as EF cathodes, an external magnetic field forces random magnetic domains to align [17, 129]. This eliminates domain walls and forms a macroscopic spin-polarized surface at the solid-liquid interface [70]. On this polarized surface, most free electrons near the Fermi level share a uniform spin orientation (e.g., spin-up, ↑) [79, 128]. **Fig. 11e** displays the molecular orbitals (MOs) of different electronic states of the oxygen molecule. These figures illustrate the distribution of electrons within the molecular orbitals formed by combinations of the oxygen atom's atomic orbitals. Each figure represents a distinct electron configuration with varying spin and symmetry properties. The ³Σ_g[−] state is the ground state of the oxygen molecule, while the ¹Δ_g and ¹Σ_g⁺ states are excited states. The ground-state oxygen molecule possesses two unpaired electrons and exhibits paramagnetic behavior. An applied magnetic field can alter the spin of these unpaired electrons, thereby modifying the molecule's electronic structure and reactivity.

Following quantum spin exchange interactions (QSEI), when ground-state triplet O₂ (↑O=O↑) adsorbs on a highly spin-polarized ferromagnetic surface, the wave functions of the spin-polarized electrons on the catalyst strongly overlap with the antibonding π orbitals of O₂ [128, 129]. Constrained by angular momentum conservation, the spin-polarized surface acts as both a spin filter and a spin injector [119]. This spin-polarized interface may favor the transfer of spin-compatible electrons to adsorbed O₂ and thereby lower the spin-

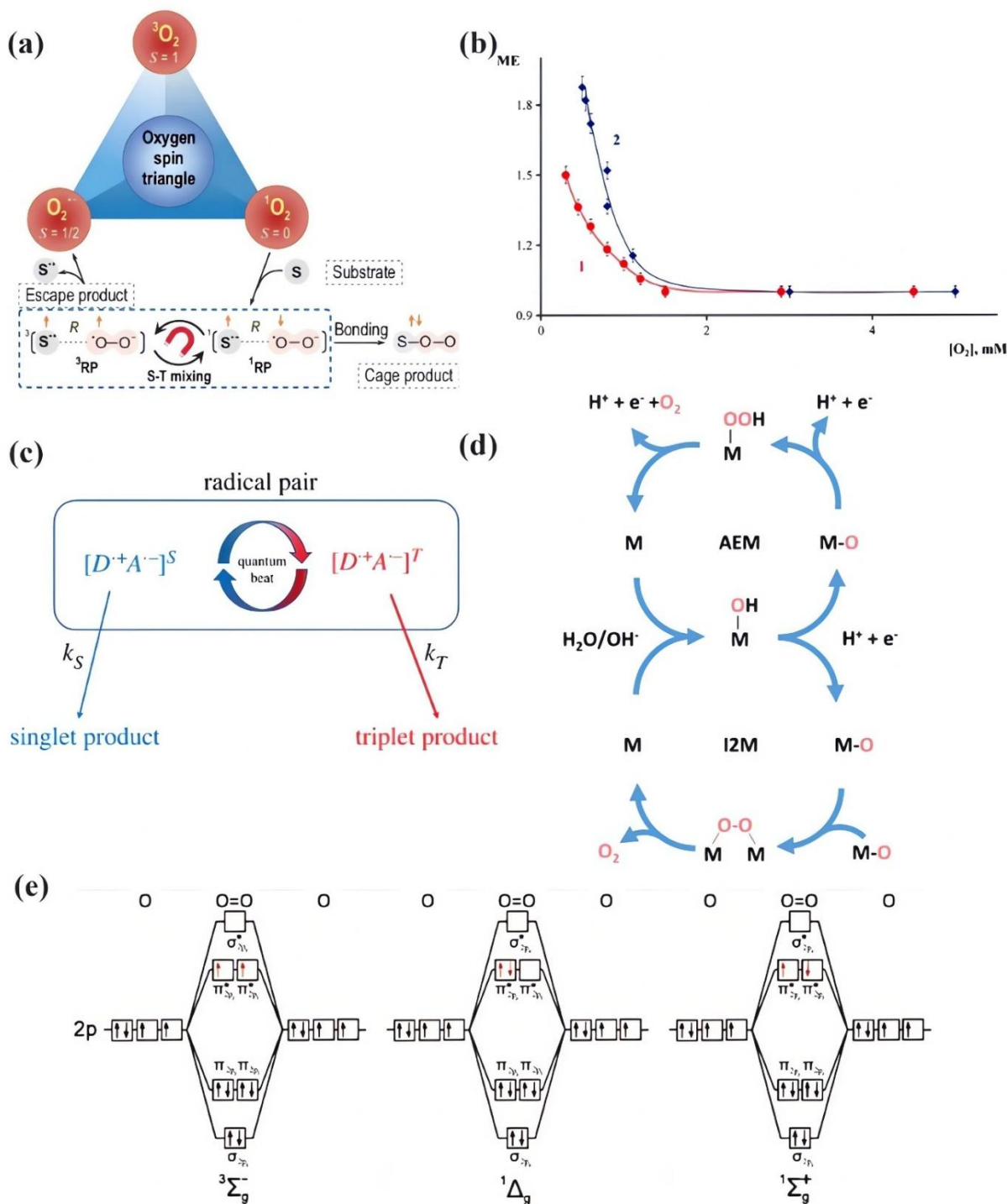


Fig. 11. (a) Schematic diagram of the oxygen spin triangle and the radical pair reaction mechanism. Modified with permission ^[130]. Copyright 2024, OUP. (b) Dependence of the magnetic effect (ME) on oxygen concentration. Modified with permission ^[131]. Copyright 2019, Wiley. (c) Schematic diagram of the spin evolution and reaction pathway of a radical pair. Modified with permission ^[132]. Copyright 2022, The Royal Society. (d) Schematic comparison of the adsorbate evolution mechanism (AEM) and the lattice oxygen-mediated mechanism (LOM) in OER, shown here only as an analogy for spin-sensitive O-O bond formation and cleavage pathways rather than as direct evidence for EF cathodic ORR. Modified with permission ^[133]. Copyright 2024, NAS. (e) Molecular orbital (MO) diagrams of different electronic states of the oxygen molecule. Modified with permission ^[134]. Copyright 2021, ACS.

related barrier for the first electron-transfer or OOH-forming step. Such a spin-selective pathway has been proposed to reduce the need for spin flipping during oxygen activation, although its direct contribution in EF cathodic ORR remains to be verified. It directly lowers the activation energy of the rate-determining step for forming the first key adsorbate (e.g., OOH)^[121]. Kinetic isotope effect (KIE) and Tafel analyses show that magnetic polarization alters the electron transfer mechanism of ferromagnetic catalysts^[89, 129]. It reduces the Tafel slope of the RDS from a traditional 120 mV/dec to below 90 mV/dec, optimizing intrinsic O₂ reduction kinetics. Additionally, because the injected electrons maintain a specific spin alignment, this mechanism blocks the parallel-spin reaction pathway that causes irreversible O-O bond cleavage^[119-121]. This mechanism may help explain improved H₂O₂ selectivity observed in some magnetic or spin-polarized catalytic systems, but it should not be regarded as direct proof of spin-controlled selectivity in EF cathodes without additional spin-sensitive evidence.

5.2.3. Chiral-induced spin selectivity in field-free environments

Alongside external magnetic field-driven spin chemistry, the chiral-induced spin selectivity (CISS) effect extends the four-species framework^[135, 136]. CISS provides an intrinsic spin-polarization alternative for EF cathode design, eliminating the need for external electromagnets^[137]. Electron transport through inherently chiral molecular layers or lattices (e.g., helical peptides, DNA, L-amino acid adlayers, or topological chiral crystals) is highly spin-dependent^[138]. This arises from strong spin-orbit coupling induced by the asymmetric electrical potential within the chiral structure^[135]. Consequently, the chiral structure acts as an intrinsic electron spin filter in field-free environments. Modifying the cathode surface with chiral molecules (e.g., L-methionine) breaks the spin degeneracy of electrons tunneling from the substrate to the adsorbed O₂, achieving interfacial spin polarization^[89, 138]. In situ attenuated total reflection infrared (ATR-IR) spectroscopy and kinetic isotope labeling confirm that this chiral microenvironment shifts the ORR rate-determining step. It transitions from generating a strongly bound O intermediate to a concerted two-electron transfer forming an OOH intermediate^[89].

Continuous injection of spin-aligned electrons satisfies the antiparallel spin pairing required for singlet H₂O₂ formation^[89, 99]. This suppresses O-O bond dissociation both thermodynamically and kinetically^[99, 121]. Coupling the CISS effect with weak external magnetic fields or ferromagnetic substrates

enables resonant spin injection^[135]. This presents a zero-energy-input strategy for designing EF cathodes with high H₂O₂ selectivity^[119].

5.2.4. Spin reshaping of orbital hybridization, bound magnetic polarons (BMPs), and the Pauling adsorption model

Flow spin state transitions alter the local electron spatial distribution and chemical bonding of catalysts^[11]. Magnetic fields tune the 3d orbital spin states of catalytic active centers (especially transition metal single atoms or clusters)^[119, 139]. This adjusts the O₂ adsorption configuration, breaking scaling relationship limits in electrocatalysis^[17, 140]. First, in transition metal catalysts with d-orbital electrons (e.g., Fe-N-C single-atom catalysts or Co-based materials), external magnetic fields or magnetic doping overcome the crystal field splitting energy^[17, 121]. This drives the central atom from a low-spin (LS) state to an intermediate-spin (IS) or high-spin (HS) state^[129]. Computational chemistry shows that the out-of-plane geometric displacement of the metal center correlates with spin crossover^[121]. The HS state increases the number of unpaired electrons in higher-energy 3d orbitals, shifting the d-band center upward^[121]. This alters the hybridization between the metal 3d and O₂ 2p orbitals and drives directional electron transfer from the catalyst to the oxygen antibonding orbitals^[99]. Spin-polarized d-p orbital coupling bidirectionally optimizes the binding energy of oxygenated intermediates (OOH, OH)^[140].

Second, in non-noble metal oxides with oxygen vacancies (e.g., CeO₂ nanocubes with specific exposed facets or Cr₂O₃/SiO₂ composites), localized carriers trapped by vacancy defects undergo strong exchange interactions with the spins of surrounding magnetic ions^[7]. This forms mesoscale bound magnetic polarons (BMPs)^[13]. Under an external magnetic field, the internal spins of these BMP clusters align ferromagnetically over long ranges^[141, 142]. This enhances charge hopping conductivity and forms localized spin-polarized micro-regions. These regions provide high-energy sites for the 2e⁻ ORR^[13, 143].

Finally, spin state regulation determines the O₂ adsorption configuration^[99, 119, 139]. In traditional dual-atom catalysts or multinuclear sites, orbital hybridization spontaneously converts the single-site end-on Pauling adsorption model (Pauling Model, M- η^1 -O₂) into a dinuclear bridge adsorption model (Bridge Model, M- μ , η^2 -O₂-M)^[117, 140]. The strong interaction in bridge adsorption stretches and cleaves the O-O bond, directing the reaction toward the 4e⁻ reduction or oxidase-like pathways. However, H₂O₂ accumulation is

the primary goal in EF systems [98, 119]. Magnetic polarization of the spin state (e.g., transitioning Fe sites to an intermediate-spin $S = 1$ state) reduces specific directional orbital overlap [99, 121]. This thermodynamically stabilizes the end-on Pauling adsorption configuration [121]. In this state, only one end of the O_2 molecule weakly binds to the metal center [117, 139]. Rapid injection of interfacial polarized electrons then balances the thermodynamics between intermediate desorption and further protonation [98, 99].

The comparison in Table 7 highlights the differences and internal connections among various quantum-level spin chemistry mechanisms for O_2 activation in EF systems. These spin-based regulation principles increasingly apply to oxygen-related catalytic reactions, including the ORR and the oxygen evolution reaction, and representative catalyst systems, magnetic-field conditions, and specific mechanisms are summarized in Table S6 in the Supporting Information.

Table 7. Comparison of three spin chemistry mechanisms regulating intrinsic O_2 activity in EF systems.

Spin chemistry mechanism and control factors	Core quantum physical principles	Regulation pathways for reaction kinetics and selectivity	Specific characteristics in EF H_2O_2 generation	References
Radical pair mechanism (RPM) and intersystem crossing (ISC)	Zeeman effect and hyperfine coupling synergistically regulate coherent radical pairs	Accelerates triplet-to-singlet ($T \rightarrow S$) conversion	Overcomes the spin barrier of singlet H_2O_2 ; increases $2e^-$ ORR selectivity	[49]
Catalyst spin polarization and quantum spin exchange interactions (QSEI)	Ferromagnetic materials eliminate domain walls; high-density spin-aligned electrons populate the Fermi level	Coherent exchange between polarized electrons and O_2 antibonding orbitals for concurrent two-electron injection	Reduces the Tafel slope; blocks the parallel-spin O-O cleavage pathway	[78]
d-p orbital coupling and bound magnetic polarons (BMPs)	Spin crossover transitions alter metal 3d electron occupancy; oxygen vacancies form long-range ordered magnetic polarons	Adjusts the d-band center; optimizes d-p hybridization and adsorption configurations	Stabilizes end-on Pauling adsorption; breaks scaling relationships	[7]

5.3. Coupling Relationships among O_2 , H^+ , Fe Species, and ROS under Magnetic Fields

Although the four key species have been discussed separately, their magnetic-field responses are strongly coupled in EF systems. O_2 is the primary precursor for cathodic H_2O_2 generation [98, 144], while H^+ availability determines the efficiency of the $2e^-$ ORR pathway [98]. Magnetic-field-induced MHD convection can simultaneously enhance O_2 supply and proton replenishment, thereby suppressing local alkalization and sustaining H_2O_2 production [144]. The generated H_2O_2 then reacts with Fe^{2+} to produce $\bullet OH$ [1], while Fe^{3+} reduction regenerates Fe^{2+} and controls the continuity of the Fenton cycle [16]. Therefore, any magnetic enhancement in ROS yield or pollutant degradation may originate from coupled changes in O_2 transport, H^+ microenvironment, Fe^{2+}/Fe^{3+} cycling, and radical generation, rather than from a single isolated species.

In addition, Kelvin-force enrichment may locally concentrate paramagnetic O_2 [11], Fe^{3+} [29], Fe^{2+} [29], and $O_2\bullet^-$ [68] near magnetic interfaces, further coupling oxygen activation with iron redox cycling. However, this enrichment can also accelerate local proton consumption and alter the interfacial pH [44], which in

turn affects Fe hydrolysis [56], H_2O_2 stability [57], and ROS selectivity [58]. Spin-related pathways may contribute to the selectivity and lifetime of ROS only after this transport and microenvironmental couplings have been excluded. Thus, the magnetic-field-assisted EF process should be viewed as an interconnected reaction network: O_2 supply and H^+ availability regulate H_2O_2 generation; Fe^{2+}/Fe^{3+} cycling converts H_2O_2 into ROS; ROS consumption and Fe speciation feed back to the local pH and interfacial reaction environment. This coupling perspective provides the mechanistic basis for the quantitative decoupling strategy discussed in Section 6.

Overall, magnetic-field-assisted EF should be viewed as a coupled reaction network rather than four independent species responses. O_2 supply and H^+ availability regulate H_2O_2 generation; Fe^{2+}/Fe^{3+} cycling determines ROS production; and ROS evolution further affects local pH, Fe speciation, and interfacial reactivity. Therefore, apparent magnetic enhancement may originate from MHD mass transfer, Kelvin-force enrichment, spin-dependent kinetics, or their combined effects. To improve readability and avoid over-attribution, Table 8 summarizes the key evidence, interference factors, and critical criteria for distinguishing these effects.

Table 8. Critical criteria for distinguishing MHD mass transfer, interfacial enrichment, and spin-chemistry effects in magnetic-field-

assisted EF systems.

Mechanism	Main evidence	Possible interference	Critical judgement
MHD mass transfer	Limiting current, mass-transfer coefficient, bubble imaging, local pH mapping	Forced convection, electrode geometry, bubble removal	Often dominates apparent enhancement
Kelvin-force enrichment	Local, O ₂ /Fe speciation, magnetic/non-magnetic electrode comparison	Competing ions, NOM, Fe complexation	Plausible but matrix-dependent
Spin chemistry	Residual kinetic enhancement, EPR, Mössbauer, XMCD, isotope labeling	MHD, pH shift, temperature, enrichment	Should be claimed cautiously
Data learning/modeling	Descriptor-performance correlation, contribution maps	Lack of standardized EF datasets	Future support tool, not direct proof

This critical comparison provides the mechanistic basis for the validation and quantitative decoupling strategy discussed in Section 6.

6. Toward closed-loop design of magnetic-field-assisted EF: materials, operando validation, and modeling

The preceding four-species analysis shows that magnetic fields can influence EF systems through MHD-driven mass transport, interfacial enrichment, and spin-dependent reaction kinetics. Therefore, future studies should focus on experimentally separating these coupled effects rather than broadly expanding the list of magnetic materials, operando techniques, or computational tools.

6.1. Integrated strategies for decoupling transport and spin effects: materials design, operando probes, and data learning

Reliable validation requires paired control systems. Magnetic and non-magnetic electrodes should be compared under similar surface area, conductivity, porosity, and Fe loading [34]. Hydrodynamic controls, such as forced-flow cells, microelectrodes, rotating electrodes, or (B || j) configurations, should be used to suppress or minimize Lorentz-force-driven convection [35]. Fig. 12(a, b) illustrates that porous electrodes and magnetic fields can improve O₂ supply and bubble release; however, these improvements may originate from mass transport rather than intrinsic spin chemistry [45]. Electrode architecture can strongly influence local magnetic-field gradients and interfacial

microenvironment formation [11]. Planar electrodes usually have more uniform current distribution and diffusion layers, making them suitable for decoupling MHD transport from spin-related kinetics [34]. In contrast, 3D foams and porous carbon felts contain edges, pores, and tortuous channels [68] that can distort local current lines, promote micro-MHD vortices [144], and create local hotspots [145] when magnetic particles or ferrimagnetic coatings are present [146]. These structures may enhance O₂ supply, Fe²⁺/Fe³⁺ enrichment, bubble removal, and local pH stabilization, but they may also cause non-uniform O₂ distribution, Fe precipitation, or bubble trapping. Therefore, stronger enhancement in porous electrodes should be interpreted as a coupled microenvironment effect rather than direct evidence of spin chemistry alone.

Recent progress in related catalytic and electrochemical fields provides useful design inspiration rather than direct evidence for magnetic-field-assisted EF systems. For example, defect-engineered MOFs may regulate coordination environments and ion/electron transport pathways, offering a possible reference for future EF catalyst design [147], reconstructed hydrogen-bond networks in related electrocatalytic systems may affect pH-dependent proton-transfer behavior, providing useful inspiration for probing proton microenvironment regulation in EF systems [148], edge-confined metal-oxygen-metal interfacial bridges may offer a useful reference for tuning orbital coupling and interfacial electron-transfer pathways in future EF catalyst design [149]. These related advances suggest that future magnetic-field-assisted EF catalysts should be designed by integrating defect regulation, interfacial bridge construction, proton microenvironment control, and sustainability considerations.

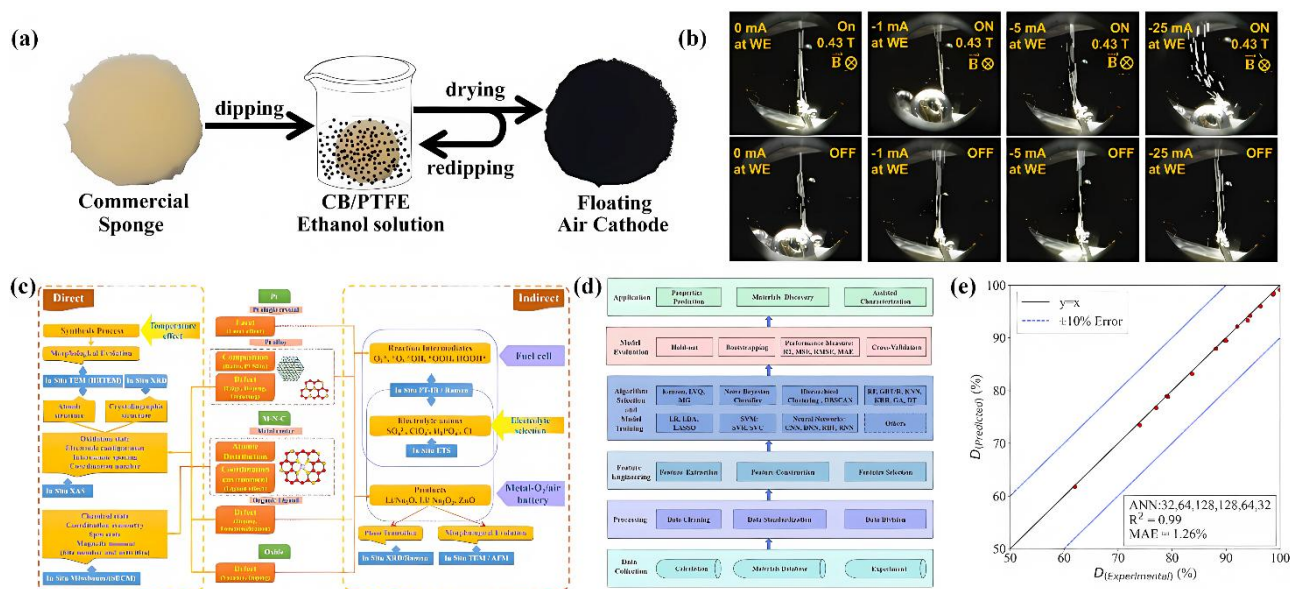


Fig. 12. (a) Fabrication of a porous floating air cathode for enhanced O_2 access and H_2O_2 generation. Modified with permission [155]. Copyright 2019, Springer Nature. (b) Magnetic-field-dependent bubble behavior near the electrode, showing possible MHD-related mass-transport effects. Modified with permission [34]. Copyright 2024, Springer Nature. (c) Machine-learning-assisted electrocatalyst discovery based on descriptors and reaction intermediates. Modified with permission [156]. Copyright 2023, Royal Society of Chemistry. (d) General data-learning workflow from data collection to model prediction. Modified with permission [157]. Copyright 2023, ACS. (e) Comparison between experimental and ANN-predicted Electro-Fenton degradation efficiency. Modified with permission [158]. Copyright 2023, MDPI.

Therefore, magnetic enhancement should only be assigned to spin-dependent kinetics after O_2 delivery, bubble removal, local pH variation, temperature rise, and Fe speciation have been independently evaluated [55].

To further standardize the interpretation of magnetic-field-assisted EF results, the magnetic-field conditions should be reported together with the electrochemical and reactor parameters [34]. Key parameters include the field type, magnetic flux density (B), field direction relative to the current ($B \perp j$ or $B \parallel j$), magnetic-field gradient (∇B^2), magnet-electrode distance, exposure time, and, for alternating magnetic fields, frequency and waveform [35–37]. These parameters may determine the dominant enhancement pathway. For example, $B \perp j$ generally strengthens Lorentz-force-driven MHD convection, whereas $B \parallel j$ can minimize macroscopic MHD and is more suitable for identifying residual spin-related kinetic effects. Non-uniform magnetic fields or magnetic catalyst surfaces can generate local ∇B^2 and promote Kelvin-force enrichment of paramagnetic O_2 and iron species [68]. In contrast, alternating magnetic fields may introduce magnetothermal effects, induced electric fields, and time-dependent spin polarization [150]. Therefore, magnetic enhancement should not be interpreted only according to magnetic-

field strength, but should be evaluated together with field direction, gradient, frequency, electrode geometry, and hydrodynamic conditions. [145] These reporting requirements are summarized in Table 9.

Although Kelvin-force enrichment can concentrate paramagnetic species such as O_2 , Fe^{2+} , Fe^{3+} , and $O_2^{\bullet-}$ near magnetic interfaces, this effect may be weakened or redirected in real wastewater matrices [11]. Background ions, including Na^+ , Ca^{2+} , Mg^{2+} , Cl^- , SO_4^{2-} , HCO_3^- , and NO_3^- , are mostly diamagnetic [23], but they can alter ionic strength, conductivity, double-layer structure, diffusion coefficients, and local MHD flow, thereby indirectly affecting magnetic enrichment [47]. Paramagnetic transition-metal ions, such as Mn^{2+} , Cu^{2+} , Ni^{2+} , Co^{2+} , and Cr^{3+} , may also compete with O_2 or Fe^{3+} in high-field-gradient regions, reducing enrichment selectivity [68]. Natural organic matter can further chelate Fe^{2+}/Fe^{3+} , form Fe-NOM colloids, block magnetic-gradient sites, and scavenge reactive oxygen species, although it may also help maintain iron solubility at near-neutral pH [84, 85]. Therefore, Kelvin-force effects should be evaluated in stepwise matrices containing representative ions and NOM, together with Fe^{2+}/Fe^{3+} speciation, dissolved O_2 , conductivity, local pH, H_2O_2 yield, and radical signals, using appropriate electrochemical and operando characterization methods

[95, 96, 151].

Table 9. Key magnetic-field parameters and their possible effects in magnetic-field-assisted EF systems.

Magnetic-field parameter	Typical condition	Possible dominant effect	Mechanistic implication
Field type	Static, gradient, pulsed, alternating	Determines whether the effect is mainly MHD, Kelvin-force enrichment, magnetothermal, or spin-related	Should be reported before assigning magnetic enhancement to a specific mechanism
Magnetic flux density, B	mT to T scale	Strengthens Lorentz force, Kelvin force, and magnetic-domain alignment	Performance may increase nonlinearly and may reach saturation
Field direction	$B \perp j$ or $B \parallel j$	$B \perp j$ enhances MHD convection; $B \parallel j$ minimizes macroscopic MHD	Useful for separating mass-transfer effects from intrinsic kinetic effects
Field gradient, ∇B^2	Uniform or non-uniform field	Non-uniform fields enrich paramagnetic O_2 , Fe species, and radical intermediates	Important for magnetic catalysts and porous electrodes
Magnet-electrode distance	Close-contact to several cm	Controls local B and ∇B^2 at the reaction interface	Affects interfacial enrichment, bubble behavior, and local microenvironment
Electrode architecture	Planar electrode, 3D foam, porous carbon felt	Alters local current distribution, magnetic gradients, and micro-MHD vortices	Stronger enhancement in porous electrodes may reflect coupled microenvironment effects
AMF frequency/waveform	Hz-kHz/MHz depending on the system	May induce magnetothermal effects, eddy currents, and time-dependent spin modulation	Temperature and non-thermal effects should be separately controlled
Exposure time	Short-term or continuous operation	Influences catalyst stability, Fe leaching, magnetic memory, and long-term performance	Needed for evaluating practical EF operation

Operando characterization should verify the real EF microenvironment under the same magnetic and electrochemical conditions used for performance tests. More specific evidence is needed to distinguish MHD transport, interfacial enrichment, and spin chemistry. MHD-dominated effects can be supported by limiting-current measurements, mass-transfer coefficients, high-speed bubble imaging, local pH mapping, and hydrodynamic controls such as forced-flow cells [34], rotating electrodes [34], microelectrodes [45], or ($B \parallel j$) configurations [55]. Interfacial enrichment should be verified by dissolved O_2 profiling [11], Fe^{2+}/Fe^{3+} speciation [29], local mapping [47], and magnetic/non-magnetic electrode controls [68]. Spin-chemistry effects should be assigned only to the residual kinetic enhancement after excluding mass transfer [32], bubble removal [34], temperature rise [35], local pH variation [45], and Kelvin-force enrichment [68], preferably supported by EPR [95], operando XAS/XES [152], Mössbauer spectroscopy [153], XMCD [154]. Therefore, pollutant degradation rate or H_2O_2 yield alone should not be used as direct evidence for spin chemistry. The data-learning workflows shown in Fig. 12c-e are retained as future tools for organizing standardized descriptors, experimental controls, evidence labels, and validated performance data, rather than as established predictive strategies for magnetic-field-assisted EF.

6.2. Theoretical calculation and mechanistic interpretation: from DFT to neural networks

Modeling should serve as supporting evidence rather than independent proof of magnetic-field-induced spin chemistry. CFD/MHD simulations can estimate velocity fields, diffusion-layer compression,

concentration polarization, bubble detachment, and O_2 delivery, while DFT or microkinetic calculations can compare adsorption configurations and reaction barriers for $O_2/ OOH/H_2O_2$ in $Fe(III)-OOH$ and $Fe(IV)=O$ intermediates [38, 86, 87, 159], ORR/H_2O_2 systems [117, 119, 160], radical/Fenton reaction networks [81, 161] and proton-transfer/ORR or data-learning modeling frameworks [162-167] under different spin states. Fig. 13a, c, d, e illustrate that magnetic-field enhancement may strongly depend on diffusion-controlled transport and local concentration redistribution, whereas Fig. 13b illustrates how theoretical calculations can help distinguish the $2e^- H_2O_2$ pathway from competing oxygen reduction pathways in ORR-related systems [162].

The key output of modeling should be a contribution map that separates mass-transport enhancement, interfacial enrichment, and residual intrinsic kinetic effects [161]. Only the residual enhancement observed after excluding MHD convection [63], bubble removal [64], local pH changes [65], and concentration enrichment [66] should be cautiously assigned to spin-dependent kinetics. The general machine-learning workflow shown in Fig. 13f is retained as an illustrative future tool for organizing descriptors, evidence labels, and validated performance data, rather than as an established predictive strategy for magnetic-field-assisted EF systems [162]. Advanced methods such as graph neural networks, Bayesian optimization, and physics-informed machine learning have shown promise in broader electrocatalysis and materials-discovery studies, but their application to magnetic-field-assisted EF remains insufficiently validated [156, 157, 162, 166]. Future datasets should include experimentally measured magnetic-field strength and direction, electrode geometry, flow

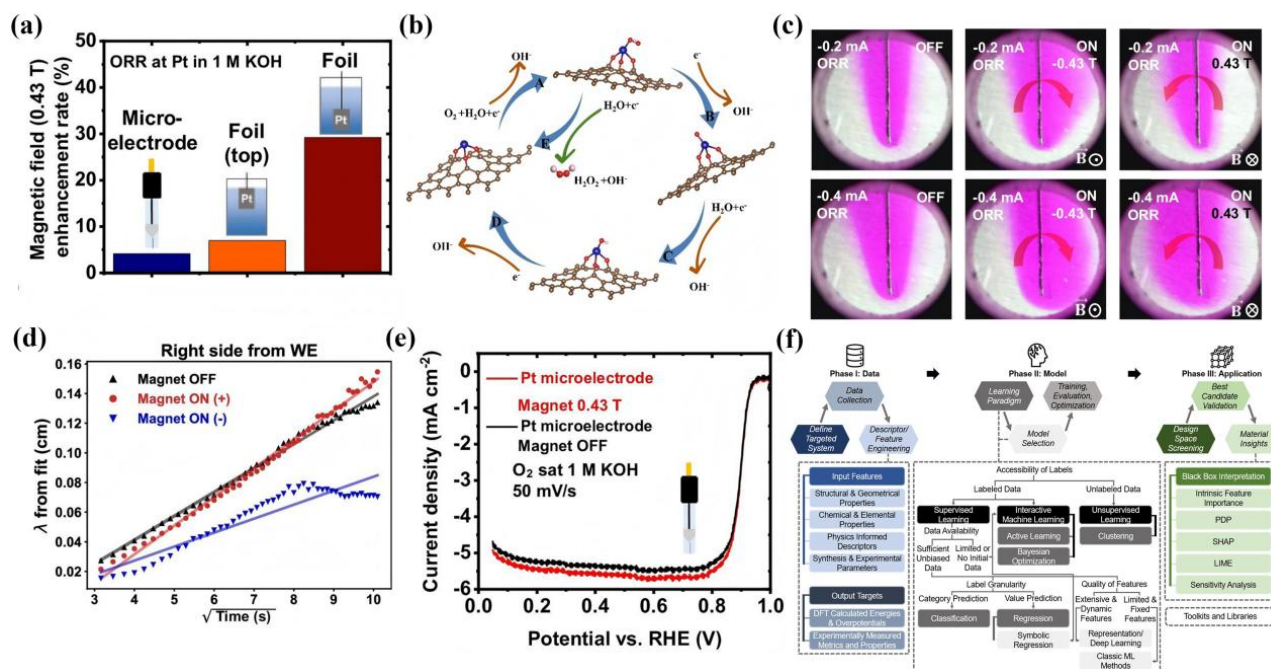


Fig. 13. (a) Magnetic field enhancement of ORR depends on electrode geometry and position, suggesting a strong transport contribution. Modified with permission [34]. Copyright 2024, Springer Nature. (b) DFT-calculated ORR intermediate cycle showing the role of OOH, O, OH, and H₂O₂-related pathways. Modified with permission [167]. Copyright 2022, Springer Nature. (c) Magnetic-field-induced redistribution of OH⁻ and change in effective diffusion behavior, visualizing interfacial concentration polarization and mass transport. Modified with permission [34]. Copyright 2024, Springer Nature. (d, e) ORR polarization curves showing that the magnetic field mainly affects the diffusion-controlled region rather than the kinetic-controlled region. Modified with permission [34]. Copyright 2024, Springer Nature. (f) ML workflow integrating descriptors, models, and catalyst screening for predictive electrocatalyst design. Modified with permission [168]. Copyright 2024, Royal Society of Chemistry.

conditions, O₂ availability, Fe²⁺/Fe³⁺ speciation, local pH, H₂O₂ yield, radical signals, wastewater matrix composition, and energy consumption. These descriptors should be supported by appropriate electrochemical and operando measurements rather than by machine-learning references alone. In this way, future materials design, operando validation, and modeling can remain focused on the central goal of this review: mechanistic decoupling of MHD transport and spin-dependent kinetics.

6.3. Quantitative estimation of MHD and spin-dependent contributions

To compare magnetic-field-induced performance enhancement across different EF outputs, a normalized enhancement factor can first be defined as:

$$E_{obs} = (P_{(B)} - P_{(0)}) / P_{(0)} \quad (5)$$

Here, P may denote an experimentally measured EF output, such as H₂O₂ production rate, H₂O₂ partial

current density, Faradaic efficiency, or pollutant degradation rate constant. This normalized factor is useful for comparing the apparent magnetic enhancement of different outputs. However, the quantitative separation of MHD mass-transfer effects from residual kinetic effects is only applicable to transport-coupled electrochemical fluxes or current-density-based outputs, such as limiting current density, H₂O₂ partial current density, O₂ reduction current, or Fe³⁺ reduction current. For Faradaic efficiency and pollutant degradation rate constants, the same normalized enhancement factor can be reported for comparison, but these outputs should not be directly decomposed using the Koutecký-Levich-type current relationship unless additional kinetic models and independent control experiments are provided.

Where P_(B) and P₍₀₎ are the performance with and without the magnetic field, respectively. The MHD contribution can be estimated from the magnetic-field-dependent mass-transfer coefficient, while keeping the intrinsic kinetic term unchanged. For O₂ reduction or

Fe³⁺ reduction, the limiting current density is:

$$j_{\text{lim}}(B) = nFk_m(B)C_b \quad (6)$$

And the measured current can be separated into kinetic and transport terms using a Koutecký-Levich-type relationship:

$$1/j(B) = 1/j_k(B) + 1/j_{\text{lim}}(B) \quad (7)$$

where $j_k(B)$ is the kinetic current density.

Experimentally, $k_m(B)$ can be obtained from limiting-current measurements using dissolved O₂, Fe³⁺, or a diffusion-controlled redox probe under identical electrode geometry [11], current density [23], magnetic-field direction [34], and flow conditions [35]. The transport-only performance under the magnetic field, ($P_{\text{MT}}(B)$), can then be calculated by replacing $k_m(0)$ with ($k_m(B)$) while assuming $j_k(B) = j_k(0)$. The relative MHD and spin-dependent contributions can be estimated as:

$$f_{\text{MHD}} = (P_{\text{MT}}(B) - P_{(0)}) / (P_{(B)} - P_{(0)}) \quad (8)$$

$$f_{\text{spin}} = (P_{(B)} - P_{\text{MT}}(B)) / (P_{(B)} - P_{(0)}) \quad (9)$$

Here, f_{MHD} represents the estimated mass-transfer contribution, whereas f_{residual} represents the residual kinetic contribution after subtracting the MHD-related enhancement. This residual term should be assigned to spin-dependent kinetics only when other magnetic-field-induced effects, including temperature rise [45], bubble removal [47], local pH variation [55], and Kelvin-force enrichment [60], have been independently corrected for or excluded.

In this framework, spin chemistry is not assigned directly from the total magnetic enhancement, but is treated as the residual kinetic contribution after subtracting MHD-related mass-transfer enhancement.

For a typical dissolved-O₂ EF cathode, $C(\text{O}_2) \approx 0.25$ mM, $D(\text{O}_2) \approx 2.0 \times 10^{-9}$ m² s⁻¹, and a diffusion-layer thickness of 50-100 μm give $k_{m,0} \approx 2-4 \times 10^{-5}$ m s⁻¹ [98]. For the 2e⁻ ORR pathway, this corresponds to $j_{\text{lim}}(0) \approx 0.10-0.19$ mA cm⁻². If the magnetic field increases k_m by 50%, the transport-only enhancement is approximately 50%. Therefore, if the observed H₂O₂ partial current increases by 70%, the estimated MHD contribution is (50/70 ≈ 71%), while the remaining (~29%) may be cautiously assigned to residual spin-dependent kinetics after excluding other interfacial effects.

The Damköhler number can further identify the controlling regime:

$$Da = j_k / j_{\text{lin}} \quad (10)$$

When $Da \geq 1$, the system is transport-controlled or under mixed kinetic/transport control, and MHD-enhanced mass transfer is expected to dominate the apparent magnetic enhancement. When $Da \leq 1$, the reaction is mainly kinetically controlled. In this case,

residual magnetic enhancement observed under control conditions that minimize Lorentz-force-driven convection, such as forced convection, microelectrodes, rotating electrodes, or parallel-field configurations ($B \parallel j$), may indicate intrinsic kinetic or spin-related contributions, but should still be verified by independent spin-sensitive or transport-control experiments [34, 35].

7. Conclusions and outlook

Magnetic-field-assisted EF technology has emerged as a promising strategy for enhancing advanced oxidation processes through the synergistic regulation of interfacial mass transport and electrochemical reaction kinetics. Unlike conventional electro-Fenton systems, external magnetic fields simultaneously influence macroscopic transport behavior and microscopic electron-spin dynamics, resulting in improved oxygen reduction, accelerated Fe²⁺/Fe³⁺ cycling, enhanced H₂O₂ production, and more efficient reactive oxygen species generation. Throughout this review, the roles of magnetic fields have been systematically summarized from the perspectives of catalyst design, magnetic-field configurations, reaction mechanisms, and practical applications.

A key contribution of this review is to introduce a four-species framework comprising Fe²⁺/Fe³⁺, H⁺, O₂, and e⁻ to systematically examine the role of external magnetic fields in the EF process. The analysis clearly separates macroscopic magnetohydrodynamic effects on mass transport from microscopic spin-chemistry effects on reaction kinetics. This separation addresses the uncertainty left unresolved in our recent review on Fe²⁺ regeneration strategies. MHD effects, driven by the Lorentz force (both bulk and micro-scale), the Kelvin force, and paramagnetic concentration-gradient forces, improve reactant delivery, accelerate the Fe²⁺/Fe³⁺ cycling, suppress interfacial pH rise, enrich O₂ at catalytic sites, and promote bubble detachment. In parallel, several microscopic pathways, including radical-pair chemistry, intersystem crossing, chiral-induced spin selectivity, catalyst spin polarization, and possible Grothuss-related proton-transfer modulation, may influence intrinsic kinetics and product selectivity under specific conditions. However, their relative contributions in EF systems still require direct operando and spin-sensitive verification. These include changes in Fe-center spin states, accelerated triplet-to-singlet evolution of radical pairs, spin-selective electron injection into triplet O₂, and stabilization of high-spin Fe(IV)=O species. More importantly, this review further translates the mechanistic decoupling framework into a

closed-loop design strategy that integrates modular material construction, operando validation, theoretical modeling, and data-driven learning.

Furthermore, recent advances in operando characterization techniques and first-principles calculations have significantly improved the mechanistic understanding of magnetic-field-assisted EF systems. Operando Raman spectroscopy, FTIR, XAS, EPR, EIS, RRDE, DEMS, and EQCM enable real-time monitoring of catalyst reconstruction, $\text{Fe}^{2+}/\text{Fe}^{3+}$ redox cycling, intermediate evolution, and interfacial reaction kinetics. Meanwhile, DFT, spin-polarized DFT, Gibbs free-energy calculations, CI-NEB, and ab initio molecular dynamics (AIMD) simulations provide atomic-scale insights into adsorption behavior, electronic structure evolution, spin regulation, reaction energetics, and dynamic catalyst/electrolyte interfaces. The combination

of advanced experimental characterization and theoretical simulations establishes a powerful framework for revealing the intrinsic mechanisms of magnetic-field-assisted electro-Fenton reactions.

Recent studies on functional materials have addressed pollutant degradation, photocatalytic H_2O_2 production, solution-plasma H_2O_2 generation, 3D-printed photocatalytic architectures, and on-site H_2O_2 electrosynthesis through the $2e^-$ ORR pathway [169–173]. These advances highlight the growing importance of integrating material design, interfacial reaction control, and oxidant generation in environmental catalysis.

Despite these advances, several important challenges remain before magnetic-field-assisted EF technology can be fully translated into practical environmental applications. Future research should focus on the following aspects (Fig. 14)

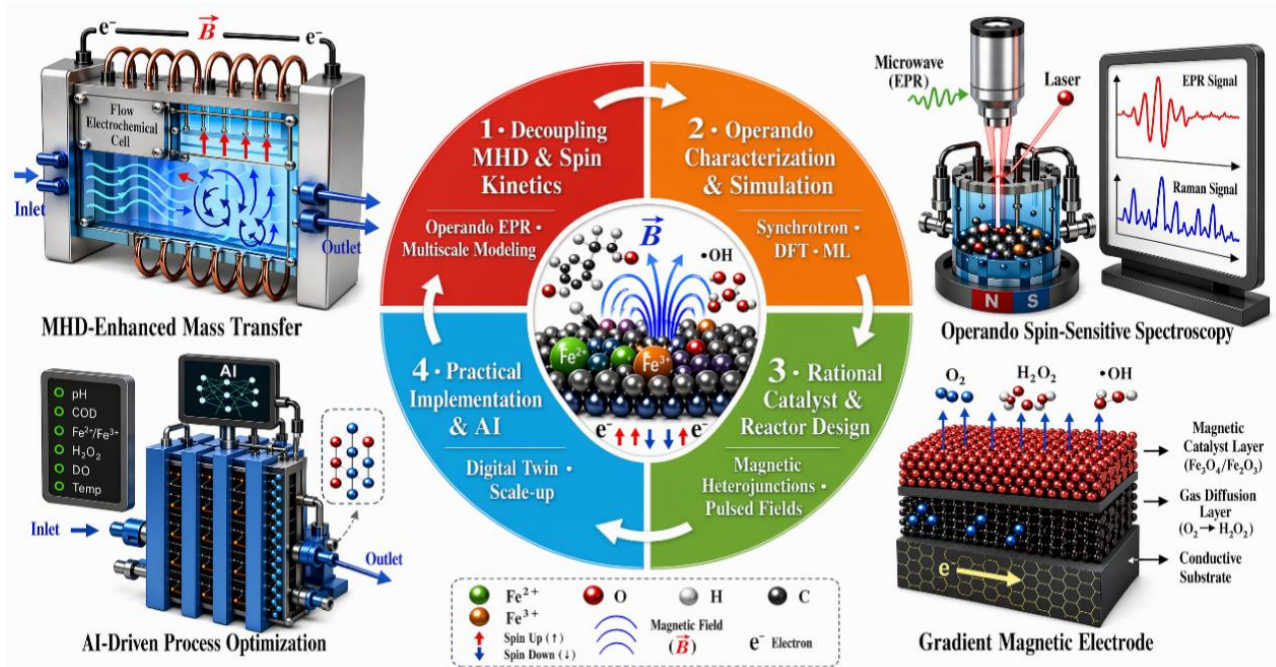


Fig. 14. Future perspectives for magnetic-field-assisted EF

(1) **Decoupling magnetic-field-induced mass-transfer enhancement from intrinsic spin-dependent kinetics.** Although both magnetohydrodynamic convection and spin chemistry contribute to catalytic enhancement, their individual contributions remain difficult to distinguish quantitatively. Future studies should combine microelectrode systems, flow-controlled electrochemical cells, and operando spin-sensitive techniques (such as EPR, Mössbauer spectroscopy, and XMCD) with multiscale simulations to establish quantitative relationships between

magnetic-field strength, interfacial transport, spin polarization, and catalytic activity.

(2) **Developing advanced operando characterization and multiscale theoretical simulations.** Most current studies still rely primarily on *ex situ* characterization, while direct operando evidence linking magnetic-field-induced catalyst reconstruction, spin polarization, $\text{Fe}^{2+}/\text{Fe}^{3+}$ cycling, H_2O_2 generation, ROS evolution, and interfacial mass transport remain scarce [96, 151, 154]. Future investigations should therefore integrate operando Raman spectroscopy, FTIR, XAS,

synchrotron-based spectroscopy, EPR, EIS, RRDE, DEMS, EQCM, and spin-sensitive probes such as Mössbauer spectroscopy and XMCD with DFT, spin-polarized DFT, AIMD, multiscale theoretical modeling, and machine-learning-assisted simulations to establish comprehensive structure-activity relationships. Such integrated experimental-theoretical approaches may provide more reliable evidence for magnetic-field-induced catalyst reconstruction, $\text{Fe}^{2+}/\text{Fe}^{3+}$ cycling, ROS evolution, interfacial electronic modulation, and spin-sensitive Fe-site evolution under realistic operating conditions, especially when in situ/operando characterization is combined with appropriate reactor controls and theoretical modeling^[96].

(3) Rational design of magnetic catalysts and reactor architectures. Future catalyst development should move beyond simply introducing magnetic components toward engineering magnetic-core/catalytic-shell structures, ferromagnetic or ferrimagnetic heterojunctions, spin-polarized active sites, and gradient magnetic-field configurations that simultaneously optimize oxygen activation, Fe redox cycling, and ROS selectivity. In parallel, reactor engineering should focus on controllable magnetic-field distributions, pulsed or alternating magnetic fields, and optimized magnetic-electrochemical coupling to maximize energy efficiency.

(4) Accelerating practical implementation and intelligent process optimization. Most current studies remain limited to laboratory-scale experiments using model pollutants. Future efforts should evaluate magnetic-field-assisted EF systems under realistic wastewater conditions containing complex inorganic ions, natural organic matter, and other matrix components, because real wastewater matrices can strongly influence EF reaction pathways and treatment efficiency^[174]. Moreover, integrating artificial intelligence, digital-twin modeling, and intelligent process control with operando monitoring may enable adaptive optimization of magnetic-field intensity, current density, and operating conditions, thereby promoting the scale-up and industrial implementation of sustainable electro-Fenton technologies.

Overall, magnetic-field-assisted electro-Fenton technology is expected to evolve from empirical magnetic-field enhancement toward mechanism-guided catalyst design and intelligent reactor engineering. By integrating advanced operando characterization, first-principles calculations, multiscale simulations, and data-driven materials discovery, a closed-loop research framework linking fundamental mechanism, catalyst design, reactor optimization, and practical application can be established. Such interdisciplinary

developments may deepen the understanding of magnetic-field-regulated electrochemical oxidation processes and promote the development of more efficient, energy-saving, and scalable EF-based technologies for environmental remediation and sustainable water treatment^[175–177].

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 article. Fengxia Deng is an Editorial Board Member of this journal and was not involved in the editorial review or the decision to publish this article.

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