

Bridging the timescales in electrochemiluminescence: From electron-transfer kinetics to rational system design

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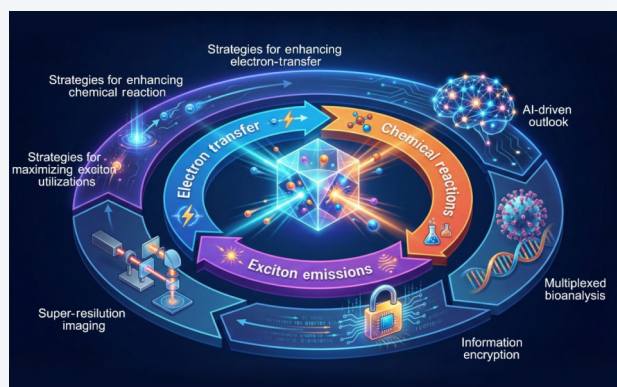
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ABSTRACT: Electrochemiluminescence (ECL) serves as a premier analytical tool, yet its performance is fundamentally constrained by the intricate interplay of physicochemical processes occurring across vast temporal and spatial scales. Herein, a coherent kinetic framework is established to deconstruct the ECL cascade into electron transfer, chemical reaction, and exciton emission modules. Building on this mechanistic foundation, we critically evaluate kinetic regulation strategies designed to overcome intrinsic efficiency bottlenecks. Furthermore, we demonstrate how these precise kinetic controls translate into advanced applications, enabling high-throughput multiplexed sensing, super-resolution imaging beyond the diffraction limit, and dynamic information encryption based on temporal logic. Finally, a prospective outlook centered on the convergence of cross-mesoscale simulation, *operando* multimodal analysis, and artificial intelligence is provided, outlining a roadmap for the development of next-generation intelligent and high-precision ECL methodologies. By integrating kinetic phenomena into a unified perspective, this work offers a conceptual blueprint for developing next-generation ECL platforms with exceptional efficiency and spatiotemporal coherence.

KEYWORDS: electrochemiluminescence, electron-transfer kinetics, electron transfer, chemical reaction, exciton emission



1 Introduction

Electrochemiluminescence (ECL) constitutes a light-emitting process wherein electrogenerated intermediates undergo electron-transfer reactions near the electrode surface to form excited states that emit photons [1–4]. Unlike photoluminescence (PL), ECL is initiated electrochemically, eliminating the need for an external light source and thereby offering minimal optical background interference [5, 6]. This unique attribute, combined with precise spatiotemporal control over the emission, has established ECL as a powerful analytical technique since its discovery in the 1960s [2]. Consequently, it serves as a cornerstone technology in clinical diagnostics [7, 8], immunoassays [9], and nucleic acid testing [10, 11].

ECL is increasingly applied in high-throughput bioanalysis,

multiplexed imaging, and dynamic information encryption, and the demands for higher signal intensity, stability, and spatiotemporal resolution have intensified [12–14]. A critical impediment, however, stems from the inherently multi-timescale nature of ECL. For example, in the classic $[\text{Ru}(\text{bpy})_3]^{2+}$ /tri-*n*-propylamine (TPRA) system [15], the emission pathway proceeds through three sequential stages [16]. First, heterogeneous electron-transfer reactions occur at the electrode interface. These are followed by homogeneous chemical reactions among the electrochemically generated intermediates, culminating in the formation of excited states. Finally, the radiative relaxation of these excitons results in photon emission. These steps correspond to distinct kinetic modules—electron transfer, chemical reaction, and exciton emission, which collectively govern the efficiency and spectral properties of the ECL output. The kinetics span an exceptionally wide range, from picosecond exciton dynamics to microsecond charge transfers and second-scale mass transport [17, 18]. The frequent mismatch among these rates leads to inefficient exciton utilization, spatial emission drift, and signal instability, ultimately limiting the performance in advanced applications [18]. Overcoming these

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limitations necessitates a holistic understanding that integrates kinetics across all relevant temporal domains. Recent pioneering studies have begun to redress this imbalance. For instance, three critical factors dictating overall ECL efficiency were identified [19], including the heterogeneous electron-transfer kinetics of the co-reactant, the stability of radical intermediates, and the spatial distribution of emitters. Through the coupling of ECL imaging with radical-trapping experiments, an efficient near-electrode reaction pathway was revealed, wherein optimized distributions of radicals and emitters yielded a remarkable 128% enhancement in signal intensity. In parallel, a shallow-trap-state strategy was introduced, employing Au–N coordination to modulate electron migration pathways in carbon nitride materials [18]. This approach effectively synchronized fast and slow processes, boosting ECL efficiency by more than fourfold. These advancements underscore kinetic regulation as a pivotal lever for ECL enhancement, transitioning the field from empirical optimization toward mechanism-guided design.

In this review, the multi-timescale kinetics and fundamental characteristics governing ECL processes are systematically delineated, encompassing the sequential stages of electron transfer, chemical reactions, and exciton emissions. Building on this mechanistic foundation, kinetic regulation strategies designed to optimize exciton generation efficiency, modulate chemical reaction rates, and accelerate interfacial charge transfer are critically evaluated. These strategies are subsequently linked to advanced applications enabled by kinetic control, demonstrating their critical role in advancing high-throughput sensing, precision bioimaging, and dynamic information encryption. Finally, a prospective outlook centered on the convergence of multiscale simulation, *operando* multimodal analysis, and artificial intelligence (AI) is provided, establishing a coherent framework to facilitate the continued evolution of intelligent and high-precision ECL methodologies.

2 Time-scale mechanistic analysis of ECL processes

2.1 Overview of basic ECL mechanisms

ECL originates from the radiative relaxation of excited states produced by high-energy electron-transfer reactions of electrogenerated intermediates [6]. Mechanistically, these processes are categorized into ion annihilation and co-reactant pathways [2, 17, 20]. Although the annihilation pathway relies on the collision of oppositely charged radical ions from the same luminophore, its application is frequently constrained by the limited stability of radical ions in aqueous electrolytes [17, 21]. Conversely, the co-reactant pathway employs a secondary species to generate energetic intermediates [19], thereby offering superior stability and efficiency in physiological environments suitable for bioanalysis. Using the prototypical $[\text{Ru}(\text{bpy})_3]^{2+}/\text{TPrA}$ system as a paradigm, the ECL emissions come from a complex sequence involving heterogeneous electrode reactions and homogeneous chemical steps (Fig. 1) [16], which exhibits a critical kinetic dependency on both interfacial electron-transfer rates and solution-phase mass transport [22].

2.2 Temporal kinetics processes map in ECL systems

The ECL process constitutes a complex kinetic network spanning from ultrafast intreelectron transitions to macroscopic mass transport, which includes the electron transfer, chemical reaction, and exciton luminescence (Fig. 2). The cascade initiates from a

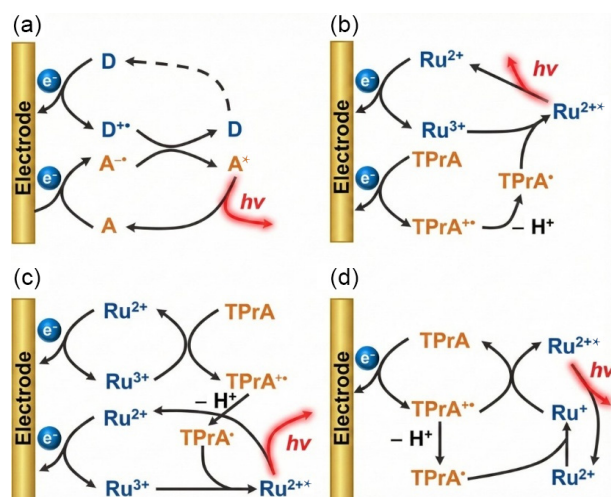


Figure 1 Schematic illustration of the ECL mechanisms in the $[\text{Ru}(\text{bpy})_3]^{2+}/\text{TPrA}$ system. (a) Annihilation route, (b) oxidative-reduction route, (c) catalytic route, and (d) low oxidation potential route.

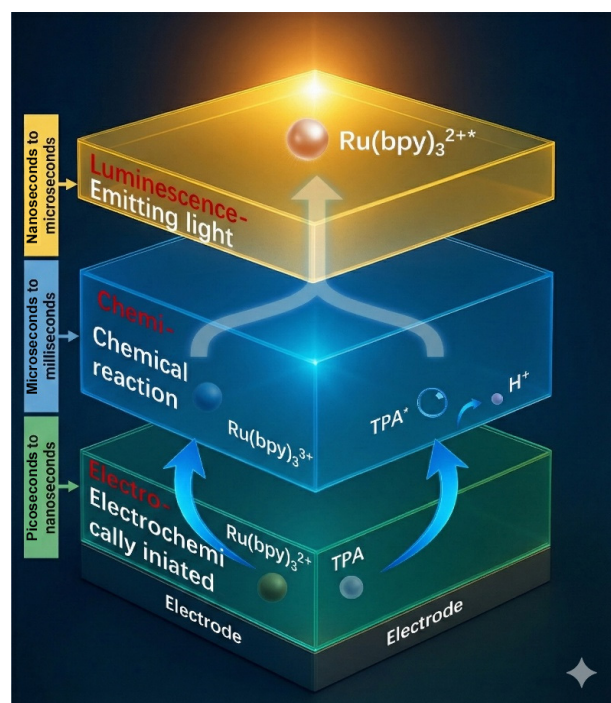


Figure 2 Typical mechanism schematic diagram of ECL.

heterogeneous electron transfer process at the electrode surface, where radical intermediates are generated [23]. Frequently, this initial step is a critical rate-limiting bottleneck, which is fundamentally constrained by the poor intrinsic conductivity of the emitter material that hinders bulk charge transport to active sites, and the high interfacial charge-transfer resistance (R_{ct}) at the electrode/emitter/electrolyte three-phase boundary [24].

After the electron injection, a chemical reaction is generally followed to produce the excited species [20, 25], which involves the homogeneous electron transfer between radical intermediates to form the excited state, and is determined by the intrinsic reactivity and stability of the radical intermediates, and the mass transport and spatiotemporal distribution of these species [26].

The final step is ECL emission that the excited ECL species relax radiatively to the ground state and release a photon. The efficiency

of this step, characterized by the luminescence quantum yield, is determined by the kinetic competition between radiative decay and various ultrafast nonradiative decay pathways, such as internal conversion, intersystem crossing, and carrier trapping [27]. The central challenge on this timescale is to engineer the emitter to minimize nonradiative decay pathways, thereby maximizing the probability of radiative decay. Different emitters face distinct and dominant nonradiative bottlenecks, necessitating tailored material-design strategies.

3 Strategies for enhancing ECL efficiency through kinetic regulation

A comprehensive understanding of multi-timescale kinetics serves as the foundation for the rational optimization of ECL systems. To overcome intrinsic limitations including high impedance, diffusion-limited reaction rates, and excitonic quenching, targeted strategies must be implemented to address specific bottlenecks at each temporal stage. These approaches can be systematically categorized into three interconnected domains encompassing enhancing electron-transfer kinetics, optimizing chemical reaction pathways, and maximizing exciton utilization (Table 1).

3.1 Strategies for enhancing electron-transfer processes

Efficiency of ECL is fundamentally governed by the kinetics of electron transfer, which initiates the generation of radical intermediates. This process, however, faces significant constraints arising principally from the high R_{ct} at the electrode–emitter junction and the limited bulk conductivity within the reticular frameworks. Consequently, enhancement strategies are designed hierarchically to first minimize interfacial injection barriers and subsequently engineer intrinsic conductive pathways within the emitter.

3.1.1 Accelerating interfacial electron injection via conductive nanohybrids

A major impediment to heterogeneous electron transfer lies in the kinetic barrier at the electrode surface, where mismatches between the electrode's Fermi level and the emitter's molecular orbitals often retard charge injection [28]. To address this challenge, a prevalent approach involves the incorporation of highly conductive nanostructures that function as “electron bridges” or kinetic relays.

The hybridization of emitters with conductive substrates, including MXenes, graphene derivatives, metallic nanoparticles (NPs), and conductive polymers, serves to expand the effective electroactive surface area while significantly minimizing the interfacial R_{ct} [29–33].

In recent years, two-dimensional (2D) nanomaterials have attracted considerable interest in this application. Specifically, MXenes are regarded as ideal substrates due to their metallic conductivity and abundant surface functional groups [29, 31, 34–36]. Recent advanced studies demonstrate that the *in-situ* growth of metal-organic frameworks (MOFs) or covalent organic frameworks (COFs) on MXenes nanosheets creates a seamless conductive interface [37]. In this configuration, the MXenes function as a rapid electron highway that facilitates electron injection from the electrode to the luminophore centers, thereby amplifying the rate of initial radical generation (Fig. 3(a)) [30]. Similarly, graphene and reduced graphene oxide aerogels provide three-dimensional (3D) conductive scaffolds [38–40]. Researchers successfully enhanced electrical contact between the emitter and the electrode by encapsulating or anchoring luminophores onto these carbon networks, effectively circumventing the insulating limitations of traditional binders (Fig. 3(b)) [41–43].

Furthermore, the integration of noble metal nanoparticles, such as AuNPs, has proven effective for mediating electron transfer [44, 45]. Beyond improving conductivity, these nanoparticles induce localized electromagnetic field effects that synergistically enhance the excitation rate [46]. Contemporary approaches utilize AuNPs assembled at the MOF/electrode interface to form tunneling junctions, which drastically reduces the overpotential required for the ECL reaction (Fig. 3(c)) [47]. Another sophisticated tactic involves engineering surface defects or modulating crystal facet exposure. Creating unsaturated metal sites or specific defect vacancies at the surface optimizes the adsorption energy of co-reactants and opens charge injection channels, resulting in superior interfacial electron transfer kinetic [31].

However, although these surface modifications significantly accelerate electron transfer kinetics, maintaining the structural integrity of the interface during continuous potential scanning remains a critical challenge for long-term applications. The repeated electrochemical cycling can induce detachment or aggregation of the conductive nanohybrids, potentially diminishing the kinetic efficiency over time. To address this, recent studies have

Table 1 Summary of kinetic regulation strategies in ECL systems

Regulatory domain	Specific strategy	Targeted kinetic bottleneck	Representative approaches/materials
3.1 Electron-transfer	Conductive nanohybrids (Section 3.1.1)	High interfacial R_{ct} and Fermi level mismatch	MXenes, graphene aerogels, AuNPs, and covalent anchoring
	Structural engineering (Section 3.1.2)	Limited intrinsic bulk conductivity and slow charge hopping	Donor–acceptor COFs/MOFs
3.2 Chemical reaction	Nanoconfinement (Section 3.2.1)	Diffusion-limited rates and instability of radical intermediates	Porous reticular frameworks and Nanoreactors
	External stimuli modulation (Section 3.2.2)	Static diffusion layers and lack of spatiotemporal control	Acoustic cavitation, potential pulses, and laser heating
	Kinetic stabilization (Section 3.3.1)	Premature non-radiative charge recombination	Stepped energy level D–A systems
3.3 Exciton utilization	Spin-state modulation (Section 3.3.2)	Spin statistics limit	TADF emitters and TTA up conversion systems
	Structural rigidification (Section 3.3.3)	Non-radiative vibrational dissipation	AIE and steric hindrance
	Spatial isolation (Section 3.3.4)	Interfacial quenching by surface defects/traps	Core–shell structures and epitaxial growth

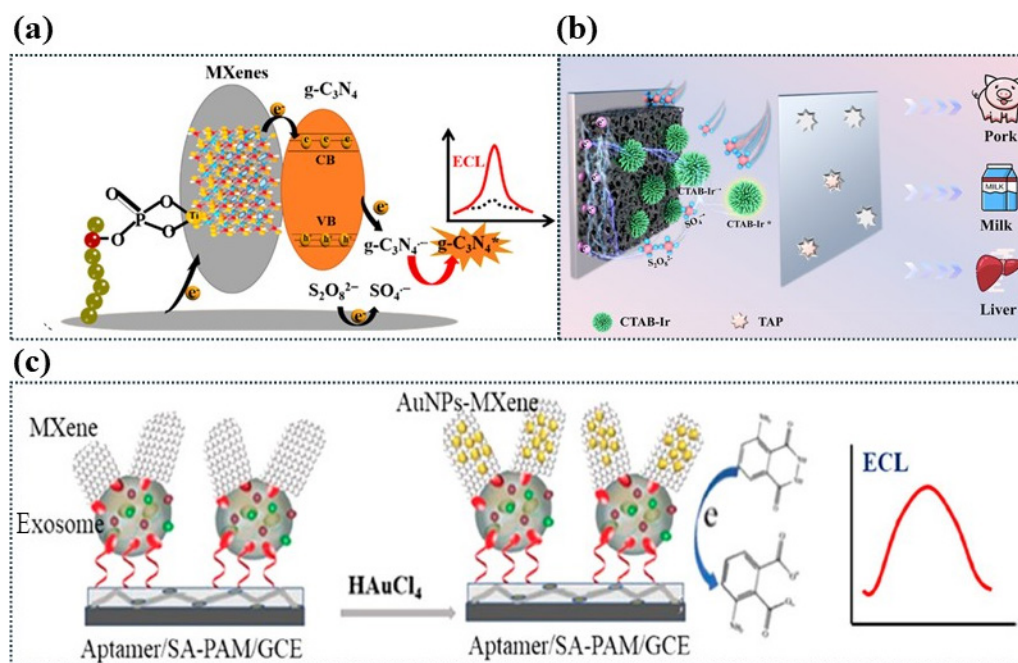


Figure 3 Mechanistic insights into accelerating interfacial electron injection via conductive nanohybrids. (a) Schematic illustration of MXenes serving as a unique conductive platform that not only accommodates g-C₃N₄ nanoparticles but also facilitates electron transfer at the electrode interface and suppresses the passivation of g-C₃N₄, enabling highly efficient and stable ECL emission. Reproduced with permission from Ref. [30], © American Chemical Society 2020. (b) Schematic illustration of a graphene aerogel with a 3D porous structure and excellent conductivity acting as an ideal carrier for AIE molecules and a multifunctional microreactor. Reproduced with permission from Ref. [43], © American Chemical Society 2025. (c) Schematic illustration of AuNPs effectively mediating electron transfer. Reproduced with permission from Ref. [45], © American Chemical Society 2020.

demonstrated that strengthening the interface via covalent bonding between the luminophore and the conductive substrate, or encapsulating the nanohybrids within protective porous carbon shells, can effectively enhance the operational stability [48, 49]. These strategies ensure that the “kinetic regulation” remains efficient and robust under sustained ECL operation

3.1.2 Enhancing bulk conductivity via structural engineering

Following efficient interfacial injection, the transport of electrons through the bulk material becomes the critical step. To mitigate the kinetic bottleneck caused by the poor intrinsic conductivity typical of porous coordination polymers, strategies focus on establishing long-range, ordered electron-transport channels [1]. This is effectively achieved by engineering donor–acceptor (D–A) conjugated architectures within the reticular frameworks [50].

By exploiting the intra-reticular charge transfer (IRCT) pathways inherent in these topologies, the typically insulating backbone is transformed into a semiconductor (Fig. 4(a)) [51]. For example, the incorporation of electron-rich ligands (donors) and electron-deficient metal nodes or linkers (acceptors) promotes p-electron delocalization across the framework [52]. Such structural continuity significantly lowers the bandgap and improves orbital overlap, thereby accelerating bulk charge mobility. This “conductive skeleton” facilitates the rapid hopping of surface-injected electrons to deep-lying luminophores within the crystal lattice, ensuring that the entire material volume participates actively in the ECL process (Figs. 4(b) and 4(c)) [51].

3.2 Strategies for enhancing chemical reaction processes

Following the initial electron injection, the generation of excited states relies fundamentally on subsequent chemical steps involving the diffusion and collision of electrogenerated radicals. However,

this stage is frequently impeded by intrinsic kinetic constraints, most notably the sluggish mass transport of co-reactants and the susceptibility of high-energy radicals to parasitic side reactions within the bulk solution. Consequently, strategies to optimize this phase focus on regulating the reaction microenvironment to stabilize intermediates and actively modulating reaction kinetics to accelerate radical annihilation [53].

3.2.1 Regulating reaction kinetics via nanoconfinement effects

To circumvent kinetic limitations imposed by slow diffusion, a paramount strategy involves the confinement of ECL reactions within restricted nanospaces [54]. Porous reticular frameworks function as catalytic nanoreactors in which the unique confinement effect significantly enriches the local concentration of reactants while stabilizing intermediate radicals against quenching. This spatial proximity facilitates a localized “closed-loop” reaction pathway that drastically lowers the activation energy for radical annihilation, thereby demonstrating kinetic superiority over stochastic diffusion in free solution (Fig. 5) [55, 56].

3.2.2 Spatiotemporal modulation of reaction flux via external stimuli

Although nanoconfinement offers a passive regulation mechanism, the reaction flux can be further amplified and controlled through active modulation utilizing external stimuli. Techniques including acoustic cavitation effectively disrupt the static diffusion layer, enhancing mass transport kinetics and promoting the mixing of reactants [57]. Concurrently, the electrochemical flux tuning permits the precise spatial manipulation of the reaction plane by directing radical collisions to specific zones (Fig. 6(a)) [58]. Furthermore, the laser-induced thermal activation enables the spatiotemporal patterning of reaction kinetics. This approach

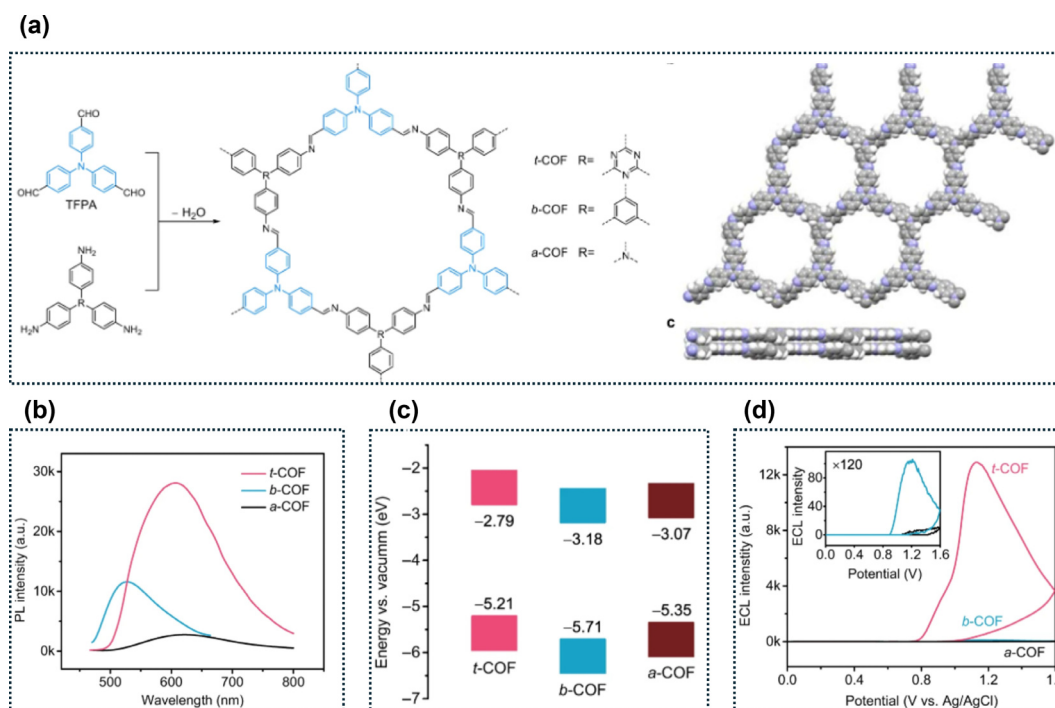


Figure 4 Mechanistic insights into enhancing bulk conductivity via structural engineering. (a) Schematic illustration of synthesis of three TFPA-based COFs. (b) PL spectra and (c) energy levels of t-COF, b-COF, and a-COF. (d) ECL curves of three COFs modified GCEs in 0.10 M PBS (pH = 6.8) contain 20 mM TPRA. Reproduced with permission from Ref. [51], © Luo, R. G. et al. 2021.

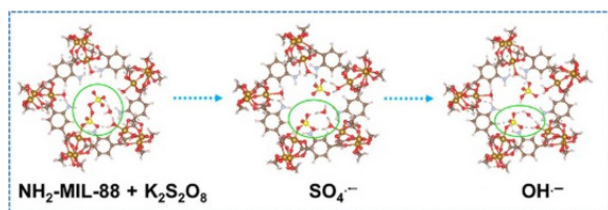


Figure 5 The adsorption model illustrating the catalysis of $K_2S_2O_8$ into free radicals on the nanoconfined NH_2 -MIL-88. Reproduced with permission from Ref. [55], © Wiley-VCH GmbH 2025.

provides a “write-erase” capability that transforms the diffusion-reaction layer from a passive matrix into a controllable parameter for on-demand ECL generation (Figs. 6(b) and 6(c)) [59].

3.3 Strategies for maximizing exciton utilizations

The culmination of the ECL cascade is the radiative relaxation of the excited state, a process fundamentally governed by the kinetic competition between the radiative decay rate and various non-radiative decay pathways [60, 61]. Consequently, the ultimate quantum efficiency of the system relies on systematically identifying and suppressing specific energy-loss channels. Strategies in this domain are hierarchically designed to address thermodynamic and kinetic bottlenecks throughout the exciton lifecycle, proceeding from the initial stabilization of charge carriers and spin-state manipulation to the suppression of intramolecular vibrational dissipation and interfacial quenching. This hierarchical optimization ensures that every generated electron-hole pair contributes effectively to the final photon output through precise kinetic steering.

3.3.1 Preventing premature recombination via kinetic stabilization

The first fundamental obstacle to exciton formation lies in the

thermodynamic instability of the precursor charge-separated state. Upon the initial electron transfer between donor and acceptor moieties, the resulting hole and electron are prone to immediate, non-radiative back-electron transfer before a stable emissive exciton can form. To overcome this kinetic instability, a prevalent strategy involves the engineering of directional charge-transfer gradients. By constructing stepped energy levels that drive unidirectional charge separation on the picosecond timescale, the system can kinetically “outrun” the rapid non-radiative recombination pathways. This mechanism ensures the longevity of the charge-separated state, thereby maximizing the yield of productive excitons, as demonstrated in sophisticated donor-acceptor “ratchet” systems [62, 63]. By engineering, stepped energy levels step up to drive unidirectional charge separation on the picosecond timescale (Fig. 7(a)). This approach kinetically “outruns” the competing rapid non-radiative recombination. This kinetic dominance ensures that the charge-separated state survives long enough to undergo productive radiative decay, significantly amplifying the ECL signal [63]. This demonstrates how manipulating the kinetics of charge separation on the picosecond scale effectively dictates the thermodynamic fate of the exciton.

3.3.2 Overcoming spin statistics limits via spin-state modulation

Once the exciton is formed, the system encounters a quantum mechanical bottleneck imposed by spin statistics, which dictates that 75% of the generated excitons populate non-emissive triplet states (T_1), leaving only 25% as emissive singlets (S_1) (Fig. 7(b)) [64]. Addressing this intrinsic efficiency ceiling requires the retrieval of dark triplet excitons into the radiative channel. Strategies such as minimizing the singlet-triplet energy gap or facilitating triplet-triplet fusion are employed to enable the kinetic upconversion of dark triplets [65]. Processes including thermally activated delayed fluorescence (TADF) and triplet-triplet annihilation (TTA) serve as

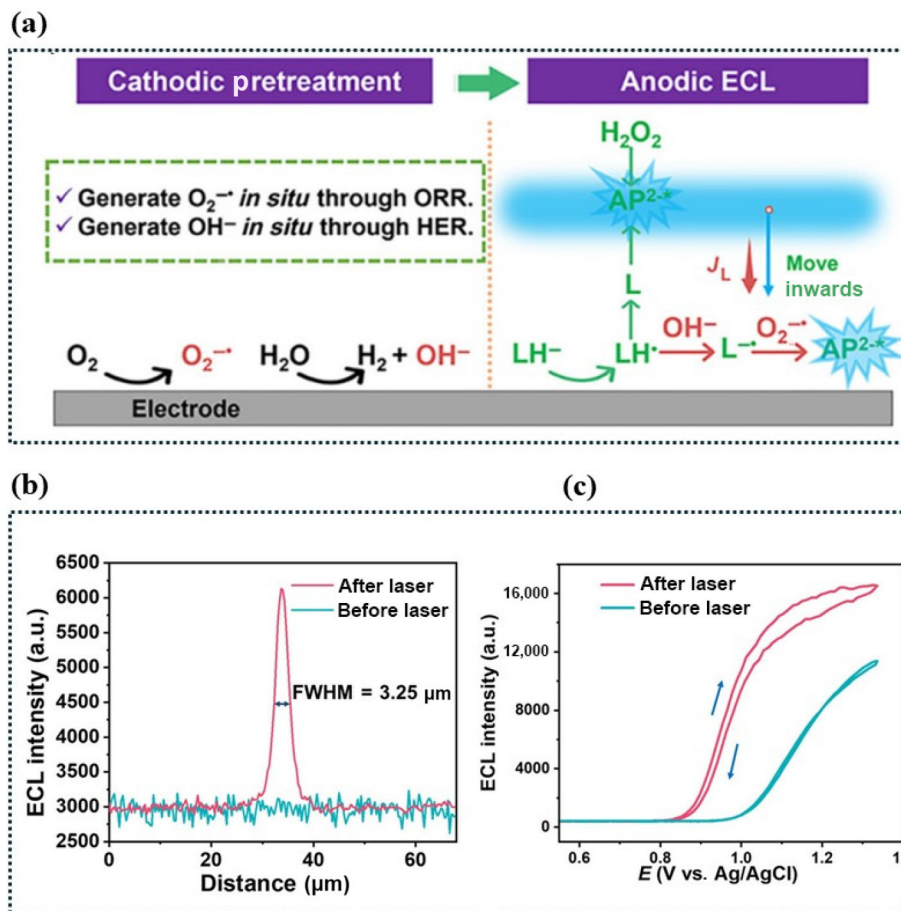


Figure 6 Mechanistic insights into enhanced ECL efficiency via spatiotemporal modulation of reaction flux via external stimuli. (a) The schematic illustration of the regulation mechanism of cathodic pre-treatment on the homogeneous reactions and the axial position of the ECL focal plane. Reproduced with permission from Ref. [58], © Wiley-VCH GmbH 2025. (b) ECL intensity profile before and after laser. (c) ECL-potential curves before and after laser. Reproduced with permission from Ref. [59], © Wiley-VCH GmbH 2025.

effective mechanisms to bypass the spin-forbidden transition, theoretically elevating the exciton utilization efficiency to near-unity [66]. Consequently, by kinetically accelerating these spin-flip processes, the system effectively circumvents the thermodynamic limit of singlet production.

3.3.3 Minimizing vibrational dissipation via structural rigidification

Following the population of the emissive singlet state, a predominant non-radiative decay channel arises from the flexibility of the molecular skeleton. Low-frequency intramolecular motions, such as bond rotations and vibrations, provide pathways for the excited state to dissipate energy non-radiatively via internal conversion [67]. The solution to this energy loss lies in the mechanical restriction of intramolecular motion (RIM) [68–70]. By inducing a rigidified environment, typically achieved through strong intermolecular steric hindrance in aggregated states, the physical motion of the emitter is locked. This structural blockade effectively shuts down the vibration-induced decay channel, a phenomenon canonically verified in aggregation-induced emission (AIE) systems where intense luminescence is activated by the suppression of conformational relaxation [71].

3.3.4 Eliminating interfacial quenching via spatial isolation

In the final stage of radiative relaxation, particularly for nanostructured emitters, the high surface-to-volume ratio

introduces a critical bottleneck: surface defects. These unsaturated coordination sites or dangling bonds function as deep trap states that capture diffusing excitons and facilitate non-radiative recombination [18, 72]. To mitigate this environmentally induced quenching, the strategy of spatial isolation of the exciton wavefunction is adopted. By epitaxially growing a wide-bandgap shell around the emissive core, the excitons are confined within the interior and physically separated from surface vacancies. This structural passivation effectively eliminates surface-mediated quenching pathways, ensuring that the relaxation of the excited state proceeds predominantly through the radiative channel (Fig. 7(c)) [18].

4 Advanced applications enabled by kinetic regulation

The rational regulation of multi-timescale kinetics, as elucidated in the preceding sections, serves not merely as a fundamental exploration but as the primary catalyst for a new generation of advanced ECL applications. By transcending simple intensity measurements, these applications leverage the precise manipulation of electron transfer, radical evolution, and exciton relaxation to encode information across temporal, spatial, and spectral domains [73]. This capability addresses critical challenges in analytical specificity, information security, and microscopic resolution.

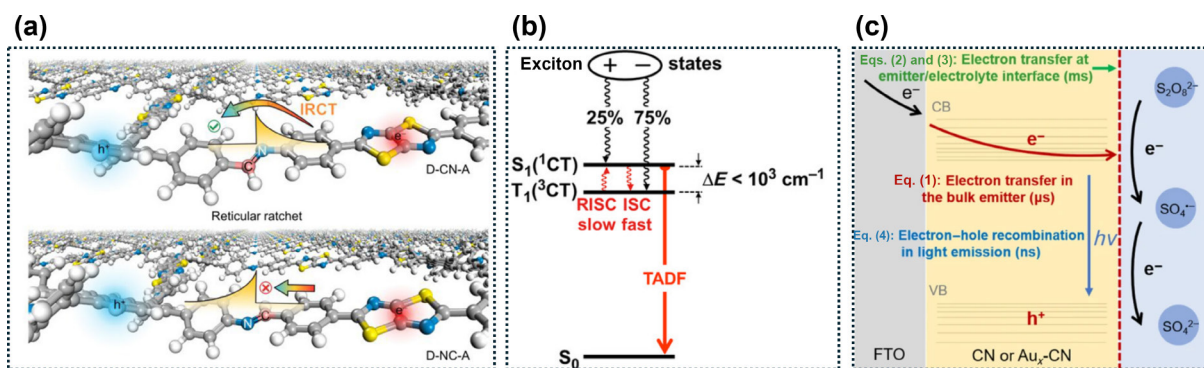


Figure 7 Mechanistic insights into enhanced ECL efficiency via maximizing exciton utilizations. (a) Schematic illustration of the charge transfer direction during electron-hole recombination in the COF structures and the corresponding ratchet effect. Reproduced with permission from Ref. [62], © American Chemical Society 2024. (b) TADF singlet harvesting. Simplified energy-level diagram to characterize the singlet-harvesting mechanism based on the TADF process. Reproduced with permission from Ref. [64], © Yersin, H. et al. 2024. (c) Schematic diagram of the Au-N bonding activated new shallow electron trap states, which worked as an electron sink in coordinating the timescale differences between the slow redox reaction at diffusion layers and fast electron transfer in bulk carbon nitrides (CN). Reproduced with permission from Ref. [18], © Fang, Y. F. et al. 2024.

4.1 Multiplexed bioanalysis via kinetic and spectral orthogonality

A critical bottleneck in biomedical diagnostics is the simultaneous quantification of multiple analytes within a single sample volume without signal cross-talk [74]. Addressing this challenge requires high-throughput screening capabilities that rely on establishing orthogonality in signal generation.

To achieve this, precise kinetic synchronization between luminophore excitation and co-reactant generation is employed. For instance, a sophisticated six-plex ECL immunoassay platform has been demonstrated for the simultaneous screening of six distinct autoantibodies within single sample wells [75]. This high-throughput capacity necessitates the rigorous modulation of electron-transfer rates (strategy 3.1) to ensure that multiple signal pathways operate without mutual interference. Furthermore, expanding detection into the spectral domain provides another vector for resolution. By engineering nanoscale probes with distinct excitonic energy levels (strategy 3.3), researchers constructed the systems capable of emitting at discrete wavelengths. This spectral orthogonality allows for the simultaneous differentiation of two antigens, effectively overcoming the limitations of monochromatic readout (Fig. 8(a)) [76]. This paradigm extends to the intelligent, ratiometric sensing where the signal accuracy is paramount. By integrating the machine-learning-assisted prediction with metal-organic frameworks (e.g., Fe-MOF) acting as electron-transfer regulators, Yuan et al. achieved dual-signal encoding from a single luminophore [77]. This approach precisely controls cathodic and anodic luminescence pathways, thereby producing a self-calibrating biosensor that is robust against environmental fluctuations (Fig. 8(b)). Consequently, the realization of high-fidelity multiplexing relies fundamentally on the precise kinetic orchestration of electron transfer and exciton recombination, which ensures signal orthogonality and eliminates cross-talk in complex biological matrices.

4.2 Dynamic information security via temporal encryption

Beyond conventional bioanalysis, the precise spatiotemporal control of ECL kinetics addresses emerging challenges in information security, particularly the vulnerability of static encryption keys to replication. The solution lies in utilizing time-

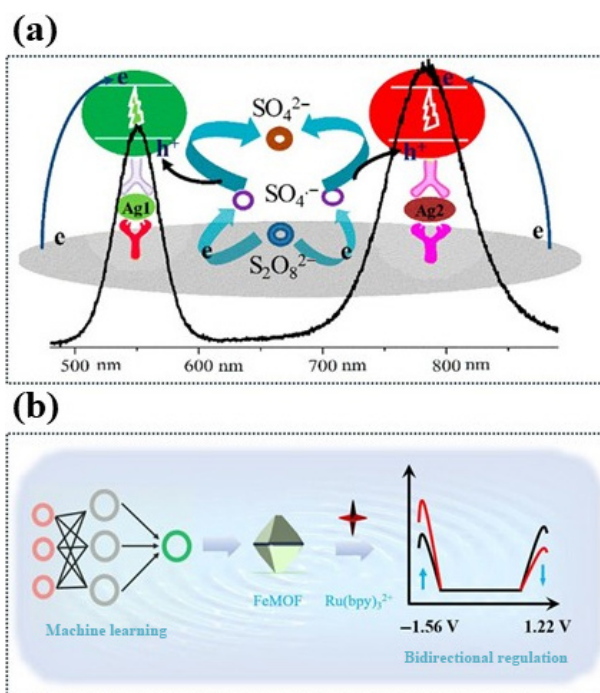


Figure 8 Multiplexed and intelligent ECL sensing platforms. (a) Schematic illustration of a spectrum-resolved dual-color ECL immunoassay designed for the simultaneous detection of carcinoembryonic antigen (CEA) and alpha fetoprotein (AFP), utilizing CdTe ($\lambda_{\max} = 776$ nm) and CdSe ($\lambda_{\max} = 550$ nm) nanocrystals as ECL tags. Reproduced with permission from Ref. [76], © American Chemical Society 2017. (b) Schematic illustration of a machine-learning-assisted strategy employing a Fe-based MOF as a bidirectional regulator to modulate the dual-emission ECL signals of a single luminophore. Reproduced with permission from Ref. [77], © American Chemical Society 2025.

resolved ECL responses as a dynamic encryption dimension that is difficult to forge.

Chen et al. [78] developed a dynamic multilevel information encryption system utilizing functional chemical inks based on iron-doped cobalt molybdate (F-CMO). Unlike strategies relying on radical lifetime modulation, this platform exploits the differential catalytic efficiency between F-CMO and undoped cobalt molybdate to generate a decisive signal gradient (spatial intensity contrast). The encryption mechanism is founded on a multidimensional security protocol comprising “biometrics (biospecific recognition),

electrochemical activation (voltage triggering), operational sequence dependence, and signal gradient judgment” (Fig. 9). By transforming abstract electrical signals into intuitive, spatiotemporally resolved visual outputs that require strict adherence to a specific decoding sequence, this approach effectively safeguards against trial-and-error decryption attempts. Crucially, the robustness of this encryption paradigm is anchored in the precise regulation of heterogeneous electron-transfer kinetics, confirming that programmable kinetic parameters can serve as high-entropy keys for advanced information security.

4.3 Super-resolution imaging via local kinetic resolution

The most profound challenge in electrochemical imaging lies in overcoming the optical diffraction limit, which fundamentally obscures microscopic heterogeneity and masks the stochastic nature of single-entity kinetics under ensemble averaging. To decipher the true structure–function relationships at the cellular and subcellular levels, ECL imaging must transcend the identification of average intensities and instead resolve local kinetic fluctuations with high spatiotemporal precision [58]. The primary strategy to breach the diffraction barrier involves the mathematical reconstruction of temporal signal variations. By analyzing the stochastic blinking or radial fluctuations of catalytic events, spatial resolution can be enhanced beyond the physical limits of the optical hardware. This principle was successfully demonstrated through the application of super-resolution radial fluctuations (SRRF) algorithms, which deconvoluted the temporal fluctuations of catalytic activity on nanostructures to reveal active site distributions with approximately 100 nm resolution [79]. Advancing toward the ultimate physical limit of quantitation, the scientific imperative shifts to single-molecule visualization. This requires not only extreme spatial confinement but also sufficient photon flux to localize individual reaction events. By constructing optical systems capable of resolving single photons, it becomes possible to directly correlate local nanoscale structure with intrinsic kinetic activity, achieving a spatial resolution as fine as 37 nm (Fig. 10(a)). This capability marks a paradigm shift from statistical inference to direct observation of single-molecule electrochemical reactions [80]. Pushing toward the ultimate physical limit, Dong et al. [81, 82] constructed an ECL microscope capable of resolving individual photons with 37 nm spatial resolution. Complementing spatial resolution, the resolution of rapid kinetic dynamics necessitates breaking the temporal

constraints of conventional imaging. Real-time observation of transient intermediates, such as those occurring at single-atom catalytic sites, demands imaging systems with millisecond-scale time resolution. Achieving this temporal precision (e.g., 100 ms) allows for the tracking of dynamic catalytic behaviors that are otherwise lost in time-averaged data (Fig. 10(b)) [83]. However, a fundamental trade-off exists between temporal resolution and the signal-to-noise ratio (SNR). Shortening the exposure time to the millisecond regime significantly limits photon accumulation per frame, which poses a severe challenge for ECL systems with intrinsically low quantum efficiency. To bridge this gap, current state-of-the-art approaches balance high-speed acquisition with adequate SNR by employing ultra-sensitive detectors, such as electron multiplying charge-coupled device (EMCCD) cameras, combined with advanced AI-assisted denoising algorithms. These technologies effectively extract valid kinetic information from photon-limited data, enabling the robust visualization of transient electrochemical events. In summary, the breach of spatiotemporal limits in ECL imaging is achievable only through the rigorous regulation of reaction kinetics at the single-molecule level, transforming stochastic fluctuations into deterministic structural information.

5 Summary and outlook

Distinct from traditional steady-state investigations, the systematic elucidation of electron-transfer kinetics across multiple timescales provides a comprehensive framework for decoding ECL dynamics, encompassing the temporal spectrum from picosecond-scale charge injection to second-scale mass transport. As delineated in the preceding sections, the integration of kinetic regulation strategies has significantly potentiated the efficiency and spatiotemporal controllability of ECL systems. Furthermore, the translation of these precise kinetic controls into advanced applications is exemplified by the realization of high-throughput multiplexed sensing, super-resolution imaging capable of circumventing the diffraction limit, and dynamic information encryption governed by multidimensional spatiotemporal logic.

The comprehensive kinetic frameworks and regulatory strategies delineated herein serve as a robust theoretical foundation for the rational design of next-generation ECL systems. By harnessing the synergy between *in silico* computational modeling and intelligent digitization, several grand challenges and transformative directions are envisioned to further elucidate intrinsic material properties and broaden application horizons.

(1) The establishment of seamless cross-mesoscale simulation frameworks, bridging quantum chemistry, molecular dynamics, and continuum models, is anticipated to enable the precise prediction of ECL responses within complex, heterogeneous environments [62].

(2) The deployment of AI-driven material discovery, utilizing machine learning algorithms and automated experimental platforms, facilitates the autonomous optimization of material composition and operational parameters, thereby accelerating the identification of ideal emitters.

(3) The development of intelligent diagnostic platforms integrated with deep learning is expected to enhance spatial resolution from the nanoscale to the molecular level, simultaneously enabling real-time image restoration [55, 84, 85].

(4) The attainment of ultrahigh spatiotemporal resolution, achieved by coupling electrochemical triggering with super-

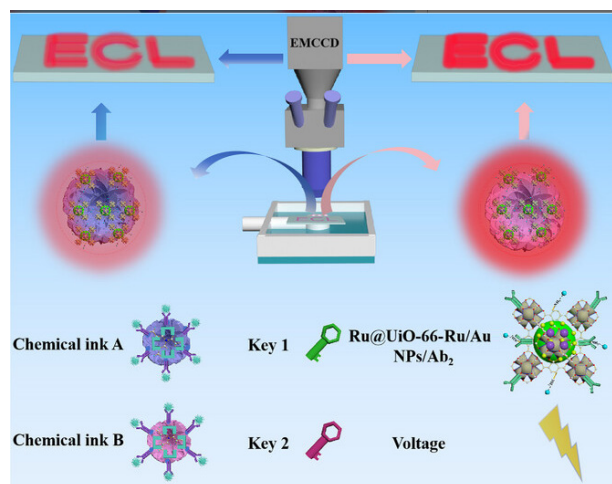


Figure 9 Chemical inks, keys, and imaging processes. Reproduced with permission from Ref. [78], © Wiley-VCH GmbH 2025.

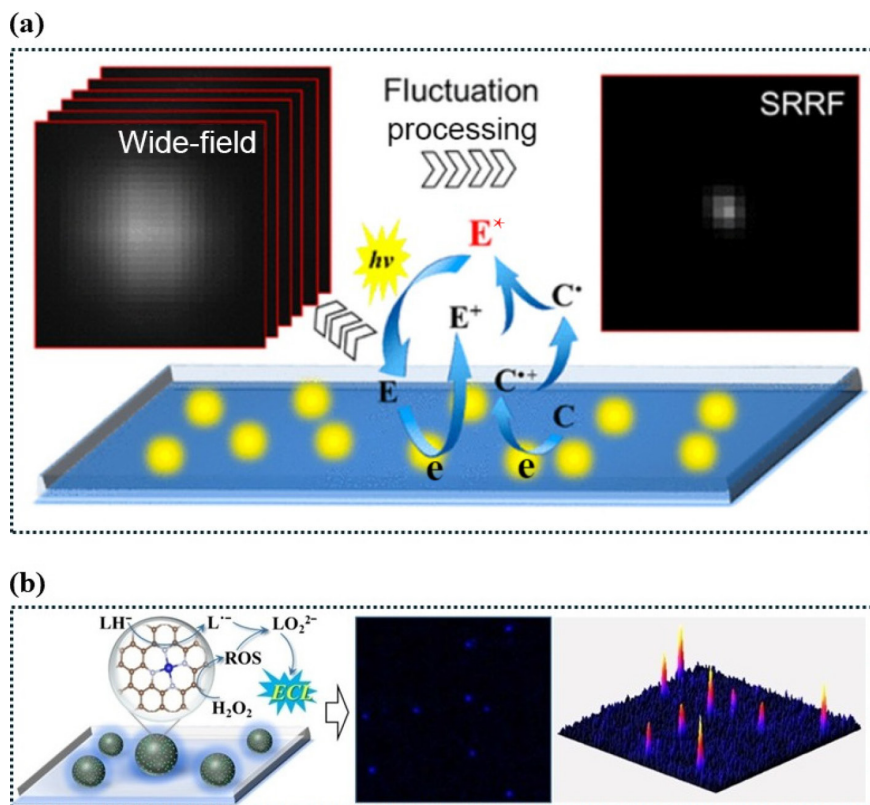


Figure 10 Super-resolution and real-time electrochemiluminescence microscopy (ECLM) for resolving single-entity kinetics. (a) Schematic of the super-resolution ECLM that could measure the facet- and site-specific activities of a single nanoparticle with nanometer resolution. Reproduced with permission from Ref. [80], © American Chemical Society 2021. (b) Schematic of the real-time ECLM to observe the electrochemical processes at individual single-atom catalysts with single photon sensitivity. Reproduced with permission from Ref. [83], © Wiley-VCH 2024.

resolution imaging, is essential for the direct visualization of single-molecule dynamics under *operando* conditions. However, capturing these rapid dynamics generates massive spatiotemporal datasets, posing a significant data processing challenge. Therefore, integrating deep learning and AI-driven analysis (as highlighted in point 2) will be indispensable for automated event recognition and trajectory tracking, thereby enabling the extraction of meaningful kinetic parameters from high-throughput imaging flows.

(5) The advancement of integrated and wearable scenarios, through the convergence of ECL with microfluidics and flexible electronics, is crucial for realizing portable, personalized healthcare monitoring.

(6) The expansion of multiscale kinetic regulation into emerging frontiers, such as *in vivo* imaging and dynamic encryption, serves to validate mechanistic robustness and diversify the technological scope of ECL.

Ultimately, the integration of multiscale kinetic regulation with advanced computational intelligence not only establishes a theoretical paradigm for decoding fundamental electron-transfer processes but also paves the way for intelligent, high-precision ECL methodologies in life sciences and information security.

Data availability

Not applicable.

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Declaration of competing interest

All the contributing authors report no conflict of interests in this work.

Author contribution statement

Y. C. S.: Investigation, data curation, writing – original draft. Y. L.: Conceptualization, project administration, funding acquisition, writing – review & editing. All the authors have approved the final manuscript.

Use of AI statement

None.

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