

Review

Electrochemiluminescence Sensing for Food Freshness Detection

Siting Wu¹, Juan He¹, Wen Li² and Wenling Gu^{1,*}

¹ State Key Laboratory of Green Pesticide, International Joint Research Center for Intelligent Biosensing Technology and Health, College of Chemistry, Central China Normal University, Wuhan 430079, China

² Hubei Key Laboratory of Plasma Chemistry and Advanced Materials, Hubei Engineering Technology Research Center of Optoelectronic and New Energy Materials, Wuhan Institute of Technology, Wuhan 430205, China

* Correspondence: wlg@ccnu.edu.cn

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Abstract: Food freshness is a critical indicator for assessing product quality and safety, intricately linked to nutritional value, sensory attributes, and consumer confidence. As global food supply chains expand and consumption patterns evolve, the demand for rapid and accurate food freshness assessment has become increasingly urgent. To address these challenges, recent research has turned to electrochemiluminescence (ECL) as an alternative detection strategy. ECL generates excited species through electrochemical reactions, offering distinct advantages in food analysis, including high sensitivity, low background noise, and straightforward visual readout. Although the broader applications of ECL-based sensing have been extensively reviewed, its specific utility in food freshness evaluation has received comparatively little attention. In this review, we provide an overview of the latest advancements in ECL for food freshness monitoring. ECL sensors demonstrate significant potential for detecting a broad range of analytes associated with food freshness, including biogenic amines, adenosine triphosphate and its degradation products, and volatile gas molecules. We further highlight engineering strategies that enhance the sensitivity and specificity of ECL-based detection technologies. Finally, the challenges and prospects for using ECL in food freshness detection are discussed.

Keywords: electrochemiluminescence; food freshness; biomarker detection; food safety

1. Introduction

Food freshness is a key determinant of food quality, safety, and public health. During storage and transportation, food deterioration can lead to nutrient loss, degradation of sensory attributes, and elevated safety risks [1]. Therefore, the development of reliable, sensitive, and user-friendly methods for assessing food freshness is essential for quality control, shelf-life management, and supply-chain optimization. Although consumers often rely on sensory cues such as color, odor, and texture to evaluate freshness, these subjective assessments lack accuracy and fail to provide quantitative information. Conventional chemical analytical methods can measure indicators such as total volatile basic nitrogen (TVB-N) [2,3], pH [4,5], and peroxide value [6], but their limited sensitivity restricts the detection of early-stage spoilage-associated molecular changes. Chromatography-mass spectrometry provides high sensitivity and specificity for identifying molecular markers, including biogenic amines [7], yet its operational complexity and limited portability hinder routine or on-site use.



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Recent advances in sensor technologies, including optical sensors [8–10], electronic noses, and electronic tongues [11–13], have greatly improved the portability and practicality of on-site detection systems. However, significant challenges remain in achieving sufficient sensitivity, selectivity, and robustness against complex sample matrices. In this regard, electrochemiluminescence (ECL) sensing has gained increasing attention due to its exceptional sensitivity and fast detection kinetics [14]. Its intrinsic compatibility with real-time monitoring and microfluidic integration further highlights its potential for next-generation compact analytical devices. Benefiting from its high sensitivity, rapid response, and relatively simple instrumentation, ECL has been widely explored in sensing applications, including environmental monitoring [15], clinical diagnostics [16] and ECL imaging [17]. Yet, although prior reviews have focused on signal amplification [18,19] and luminophore engineering [20,21], the practical implementation of ECL remains insufficiently explored. In particular, its application in real-time food freshness monitoring has been largely overlooked. Therefore, the development of sensitive and accurate ECL platforms integrated with smart, portable devices is urgently needed for food freshness detection.

In this review, we present recent advances in ECL sensing for food freshness detection (Figure 1). We first introduce the underlying luminescence principles of ECL, which are intrinsically linked to its superior detection performance. Then, the latest developments in ECL-based sensors are presented. Specifically, we explore the engineering strategies of ECL sensors designed to achieve efficient detection signals. Signal amplification strategies, including resonance energy transfer, co-reactant amplification, and catalyst-assisted amplification, have been widely employed to improve sensor sensitivity, while molecular imprinting and aptamer-based techniques contribute to improved analytical selectivity. Finally, we address the current challenges and prospects associated with ECL-enabled food freshness sensing.

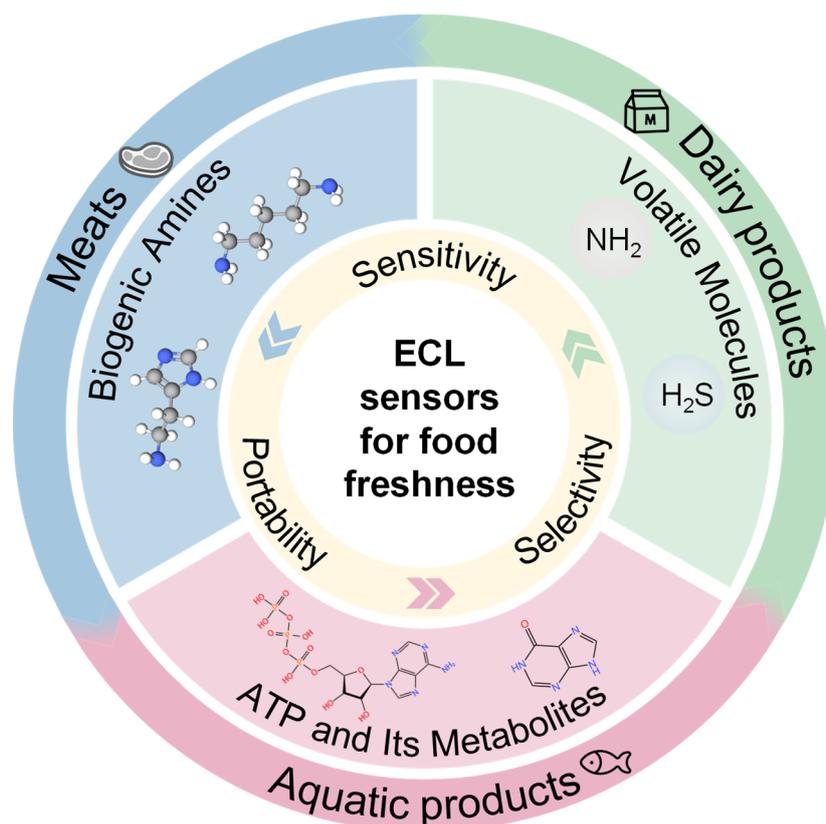


Figure 1. The scheme of the electrochemiluminescence sensing for food freshness detection.

2. ECL Mechanisms

ECL arises from the coupling of electrochemical reactions with chemiluminescent light emission. Upon application of an appropriate potential, electroactive species are generated at the electrode surface and undergo subsequent electron-transfer reactions among themselves or with other components in the system, leading to the formation of excited states that emit photons upon relaxation to the ground state [18,22–24]. As the excitation process is driven electrochemically rather than optically, ECL offers excellent controllability and is inherently free from interference by ambient light. ECL emission generally proceeds via either the annihilation mechanism or the co-reactant mechanism [25]. In the annihilation route (Figure 2A), alternating potentials generate oxidized and

reduced forms of the luminophore that recombine to produce an excited state. However, this route typically requires a wide potential window. By contrast, the co-reactant route enables efficient ECL generation under a single-direction potential scan and is therefore more widely adopted in analytical sensing. In this mechanism, both the luminophore and a co-reactant are electrochemically activated, and the electro-generated co-reactant intermediate reacts with the luminophore to produce the emissive excited state.

Notably, many freshness-related compounds, particularly biogenic amines, can directly participate in or modulate co-reactant ECL processes, thereby establishing a clear mechanistic link between ECL emission and food freshness. As illustrated in Figure 2B, in the $\text{Ru}(\text{bpy})_3^{2+}/\text{TPrA}$ system, anodic polarization triggers oxidation and deprotonation of TPrA to generate a strongly reducing radical intermediate, which reacts with Ru^{3+} to form the excited state and emit light. The magnitude of the ECL signal is closely related to the concentration of electroactive species in solution, such that increased availability of co-reactants generally leads to enhanced ECL intensity. During food spoilage, the accumulation of freshness-related biomarkers and their degradation products, such as hydrogen peroxide (H_2O_2) or dissolved oxygen (O_2) [26–30], can act as alternative co-reactants and induce measurable changes in ECL intensity. Owing to its electrochemical control, ECL exhibits excellent tunability. Compared with fluorescence-based methods, ECL can achieve significantly lower detection limits, reaching the approximately 10^{-12} – 10^{-15} mol/L range [21].

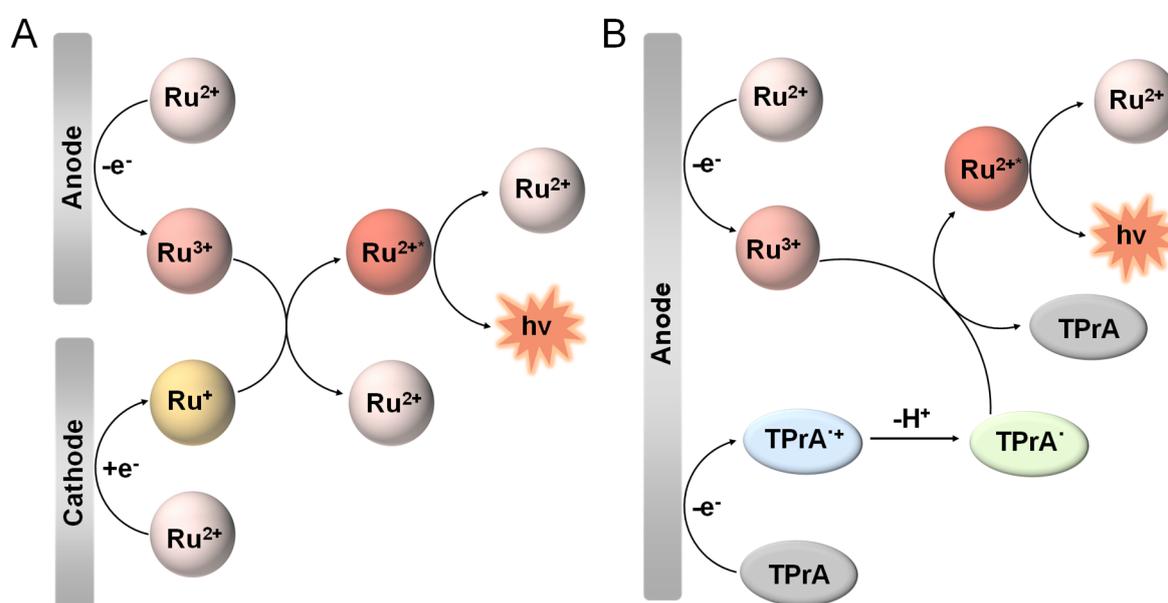


Figure 2. Schematic of the mechanisms for (A) annihilation ECL and (B) co-reactant ECL systems.

3. Detection of Food Freshness Markers

Food freshness markers are not universally applicable but are highly dependent on the specific food matrix and dominant spoilage pathways. In protein-rich foods such as meat, biogenic amines are highly responsive indicators of microbial and enzymatic degradation. ATP and its degradation products are more suitable for muscle-based foods, particularly aquatic products, where postmortem energy metabolism governs freshness changes. In contrast, volatile compounds provide rapid and non-destructive freshness information but often suffer from limited selectivity in complex systems. Therefore, freshness assessment strategies should be tailored to food-specific characteristics rather than relying on a single class of markers.

3.1. Biogenic Amines

During storage, meat undergoes extensive protein degradation driven jointly by microbial metabolism and endogenous enzymatic activity, generating a range of nitrogen-containing compounds. Among these, biogenic amines are widely recognized as representative freshness markers because their concentrations increase sensitively with the progression of spoilage [31]. Biogenic amines are produced through the decarboxylation of amino acids and can be broadly categorized into monoamines and polyamines, with histamine (HA) and tyramine being the most frequently monitored indicators due to their strong correlation with microbial activity and food safety concerns.

Conventional analytical approaches for biogenic amine determination include optical sensing [32], chromatography-mass spectrometry [33], and electrochemical sensing [34]. These techniques typically rely on signal transduction generated by the interaction between the alkaline nature of biogenic amines and specific chemical recognition elements [35]. For instance, Jastrzębska and colleagues employed chromatographic-mass spectrometric techniques to simultaneously quantify multiple biogenic amines with high resolution and accuracy [36]. Despite their excellent analytical performance, such methods require labor-intensive sample pretreatment, which limits their suitability for routine or on-site monitoring. Table 1 summarizes the detection limits and analytical sensitivities of different detection methods.

Table 1. Comparison of analytical performance of different methods for food freshness detection.

Detection Method	Target Marker	Food Matrix	Linear Range	LOD	Ref.
ECL	Tryptamine	Vinegar, soy sauce, and fresh fish	1 nM–0.1 mM	0.25 nM	[37]
	Histamine	Liquor, white vinegar, and fresh fish	5 nM–0.1 mM	1.71 nM	[38]
Chromatography	Histamine	Beef, pork, and chicken breasts	8.60–73.52 mg L ⁻¹	0.7 mg L ⁻¹	[36]
	Histamine	Pork	/	0.03 mg kg ⁻¹	[39]
Colorimetry	BAs	Fish	400–1000 μM	2.73 μM	[40]
	Histamine	Chicken meat	10–100 μg mL ⁻¹	0.21 μg mL ⁻¹	[41]
Fluorescence spectroscopy	Histamine	Fish	0.5–100 ng/mL	0.1 ng mL ⁻¹	[42]
	Histamine	Fish	50 ng–40 μg mL ⁻¹	30 ng mL ⁻¹	[43]
Electrochemical analysis	Tyramine	Fish	5–150 mg L ⁻¹	0.4 mg L ⁻¹	[44]
	Histamine	Fish	0.01–100 ng mL ⁻¹	1.25 pg mL ⁻¹	[45]

To address this limitation, ECL sensing has been increasingly explored for the rapid detection of representative biogenic amines. Senabut and colleagues utilized HA as a co-reactant in the tris(2,2'-bipyridyl)ruthenium ECL system (Figure 3A), enabling its quantification through HA-induced enhancement of ECL intensity [46]. However, other biogenic amines containing free amino groups can also enhance the ECL signal, generating nonspecific luminescence responses that may cause false positives and compromise analytical accuracy. To improve selectivity, An et al. implemented capillary electrophoresis-electrochemiluminescence technology to achieve specific recognition of HA while eliminating interference from other common amines, achieving a detection limit of 4.19×10^{-2} mg/L [47]. In parallel, molecularly imprinted polymers have been widely integrated with ECL platforms to provide selective recognition cavities tailored to specific amines by matching the size, shape, and functional groups of template molecules. This strategy has been successfully used for the selective detection of tryptamine [37], histamine [48], and tyramine [49]. Beyond molecular recognition strategies, data-driven approaches have recently been introduced to further enhance analytical accuracy. Lu and co-workers integrated a deep learning-enabled smartphone platform with a dual-potential-resolved ECL sensor, achieving highly sensitive and selective on-site detection of tyramine in various meat products (Figure 3B) [50]. By extracting multidimensional features from ECL intensity patterns and potential-resolved signals, the deep learning model effectively distinguished target-induced signals from background interference, thereby improving both selectivity and quantification accuracy under complex sample conditions. This integration highlights the potential of artificial intelligence-assisted ECL systems for robust on-site monitoring of biogenic amines in real food matrices.

3.2. ATP and Its Degradation

In early studies of food preservation and spoilage monitoring, the purine metabolic cascade involving adenosine triphosphate (ATP) was recognized as a fundamental pathway for assessing the postmortem energy depletion in biological tissues [51]. Following animal death, ATP undergoes a series of enzymatic transformations, sequentially degrading into adenosine diphosphate/adenosine monophosphate, inosine monophosphate (IMP), hypoxanthine ribonucleoside (HxR), hypoxanthine (Hx) and ultimately Uric acid [52]. Among these metabolites, Hx is particularly noteworthy because of its chemical stability and gradual accumulation during storage, making it a reliable indicator of freshness deterioration in both meat and aquatic products. Based on this metabolic cascade, Saito et al. introduced the concept of the “K value” [53], defined as the ratio of the total concentration of HxR and Hx to the total concentration of ATP and its degradation products. This parameter has been widely adopted as a standard freshness criterion in fish and seafood, with a K value exceeding 58% generally indicating spoilage [52].

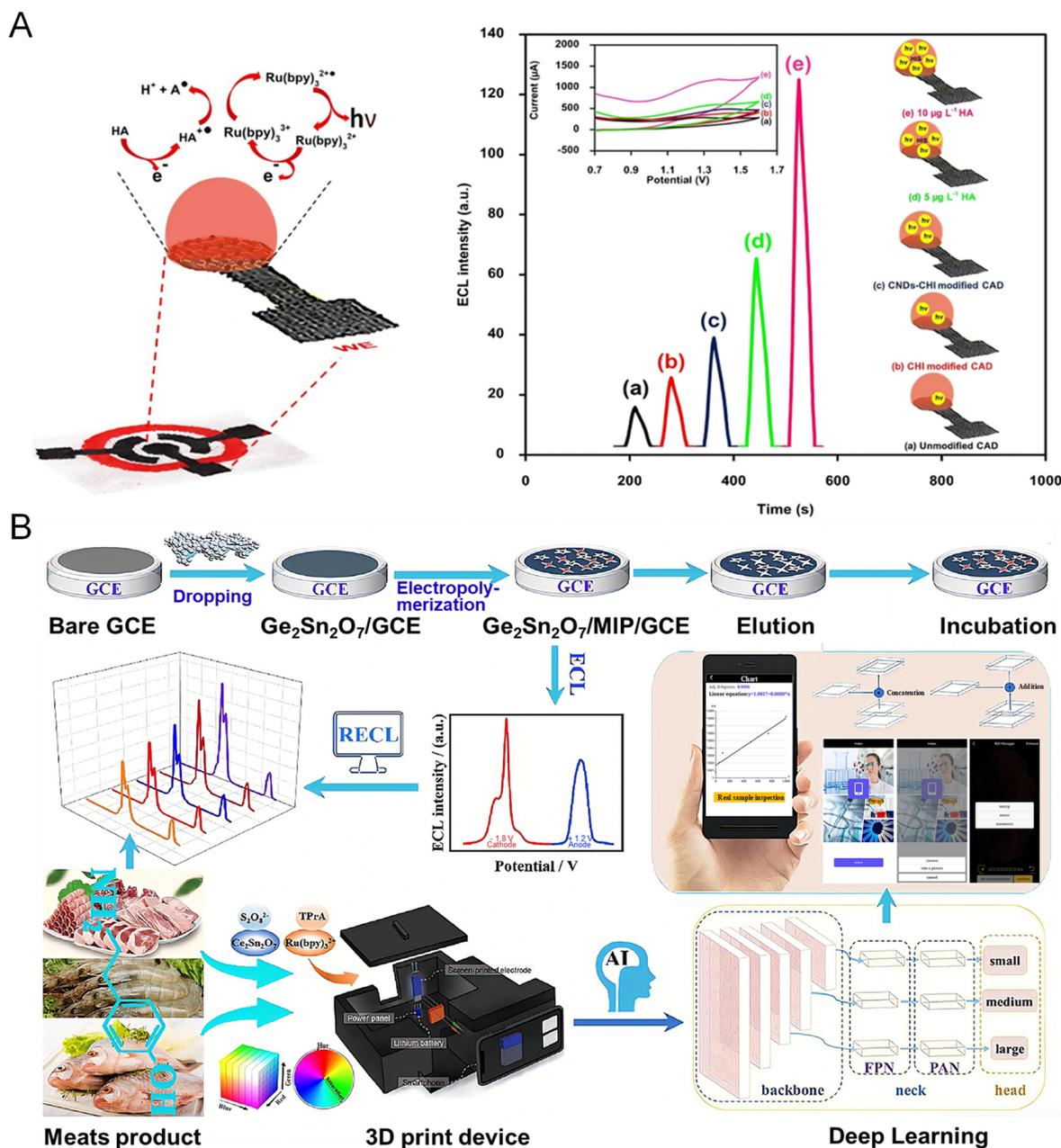


Figure 3. (A) Schematic illustration of the mechanism and ECL signal of $\text{Ru}(\text{bpy})_3^{2+}$, with HA acting as the co-reactant on the CAD-ECL platform. Reprinted with permission from Ref. [46]. Copyright 2023, Springer. (B) Schematic illustration of the construction of the fabricated machine learning-assisted smartphone-based visual and portable intelligent detection of TYM in agro-food products. Reprinted with permission from Ref. [50]. Copyright 2023, Elsevier.

Consequently, ECL technology has emerged as a promising tool for detecting HxR owing to its compatibility with enzymatic reactions. In 2008, Lin et al. pioneered an enzymatic amplification platform based on xanthine oxidase [54], in which hypoxanthine was enzymatically converted into H_2O_2 that subsequently acted as a co-reactant to enhance the luminol-based ECL emission (Figure 4A). This approach enabled the precise quantification of hypoxanthine, providing a direct analytical basis for estimating the numerator of the K value. Building on this, Wang et al. introduced a dual-enzyme cascade system capable of degrading IMP and its downstream products, with hydrogen peroxide generated at each step serving as a signal-amplifying co-reactant [55]. Such cascade designs allow simultaneous monitoring of multiple ATP degradation products, thereby improving the accuracy of K value assessment. In addition to indirect metabolite detection, ECL platforms have also been developed for direct ATP quantification. Yao and colleagues reported an ATP aptamer-based ECL signal-switching strategy (Figure 4B), in which ATP recognition triggered a conformational rearrangement that repositioned the luminophore close to the electrode surface, achieving an ultralow detection limit of 0.05 nM with high selectivity [56]. More recently, Li et al. constructed a “signal-off” ECL sensing platform based on resonance energy transfer between ATP and a

self-luminescent metal-organic framework [57]. This approach enabled stable and reproducible ATP recognition without complex interfacial engineering (Figure 4C). Collectively, these ECL-based strategies provide complementary routes for quantifying both ATP and its degradation products, thereby offering a robust analytical foundation for accurate K value determination in the freshness evaluation of aquatic products.

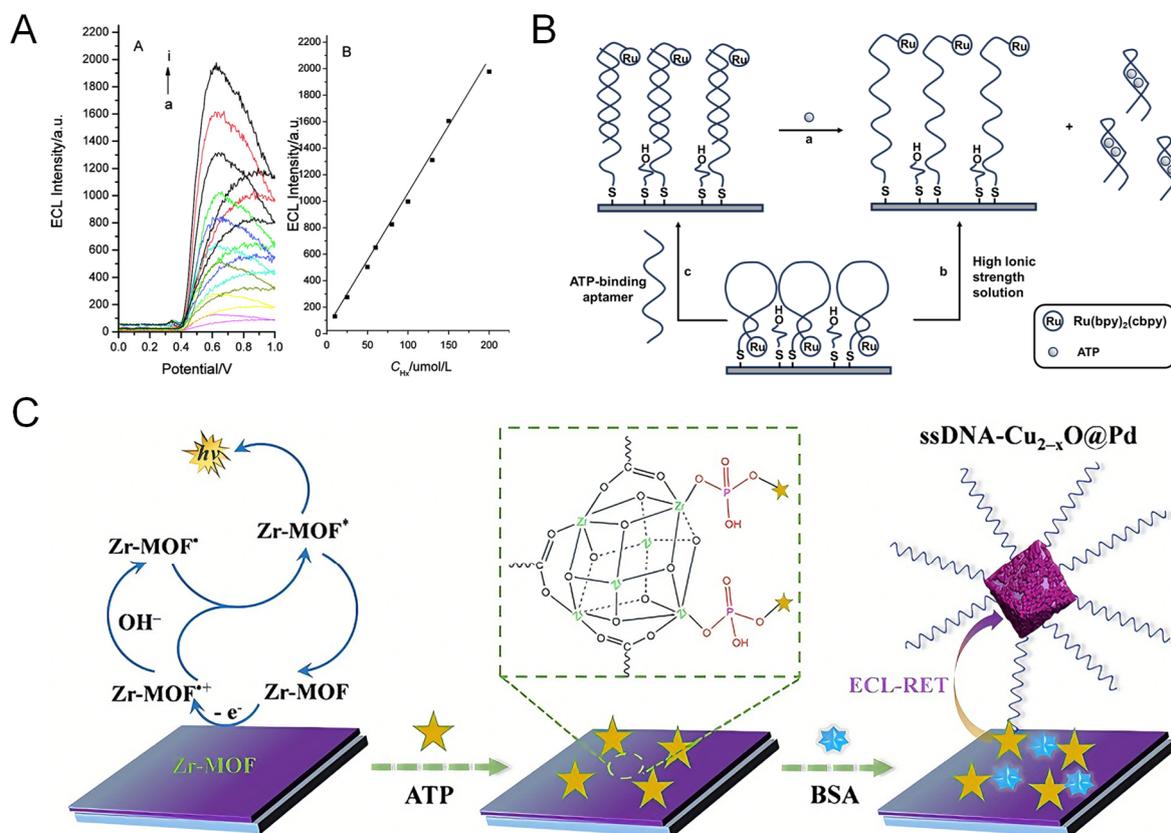


Figure 4. (A) Schematic of ECL-potential curves of luminol-Hx system on XOD/HCPE with different Hx concentrations. Reprinted with permission from Ref. [54]. Copyright 2008, American Chemical Society. (B) Schematic of the aptamer-based ECL biosensor for ATP detection and its regeneration. Reprinted with permission from Ref. [56]. Copyright 2009, Elsevier. (C) Schematic of the mechanism for ATP detection using aggregation-induced emission luminophore. Reprinted with permission from Ref. [57]. Copyright 2024, American Chemical Society.

3.3. Volatile Molecules

Volatile compounds play a crucial role in indicating the onset of spoilage and the progression of food degradation [58]. This is particularly evident in dairy products, which are highly susceptible to protein hydrolysis, lipid oxidation, and microbial growth, resulting in a progressive decline in sensory and nutritional quality. Among commonly monitored markers, TVB-N and volatile sulfur compounds provide complementary insights into spoilage progression, as both correlate with characteristic off-flavors. Their early release and rich information content have motivated the development of gas-targeted sensing strategies that enable rapid and non-destructive freshness assessment [59].

TVB-N, which is mainly composed of ammonia, dimethylamine, and trimethylamine, represents the final product of the decomposition of both protein and non-protein nitrogen. Trimethylamine (TMA), a key component of TVB-N, typically increases significantly as microbial metabolic activity intensifies [60]. Gao and colleagues developed an Fe(II)-EDTA system to reduce trimethylamine oxide to TMA, which then acted as an efficient co-reactant for Ru(bpy)₃²⁺, significantly amplifying the ECL signal [61]. This approach enabled the quantitative analysis of TMA in six different categories of aquatic products, demonstrating the utility of ECL technology in food freshness monitoring. Praoobon et al. further advanced TMA detection using a microfluidic paper-based analytical device (μ PAD)-ECL platform (Figure 5A), providing a low-cost and portable method for quantifying TMA in fish samples [62]. This platform showed excellent correlation with results obtained by benzoyl-derivatization high-performance liquid chromatography.

Building on the detection of nitrogenous volatiles like TMA, hydrogen sulfide (H_2S) has emerged as a complementary marker that reflects early microbial and oxidative processes, particularly relevant to dairy spoilage. Li and his team developed a dual ECL/fluorescence probe for ultrasensitive detection of H_2S in homogeneous media (Figure 5B) [63], achieving detection limits of 0.18 pM in ECL mode and 0.4 nM in fluorescence mode. The probe was further immobilized onto a paper-based platform, enabling rapid, on-site sulfide monitoring and linking nitrogenous and sulfurous volatiles in comprehensive spoilage assessment. However, while these liquid-phase techniques are highly sensitive and reliable, they are inherently limited by their dependence on solution-based analysis. To address the challenge of in situ detection of headspace volatiles, Torul and colleagues developed a paper-based ECL sensor capable of directly collecting gaseous ammonia [64]. This sensor utilizes an electron transfer reaction between gas-phase ammonia (NH_3) and $\text{Ru}(\text{bpy})_3^{2+}$ immobilized on the electrode surface, triggering the ECL response and enabling the direct quantification of ammonia in food samples (Figure 5C). Despite this advancement, gas-phase sensors still face challenges related to selectivity when applied to complex food matrices. Therefore, the development of ECL sensing platforms that can selectively and directly capture volatile spoilage markers in authentic food environments remains a critical challenge in food spoilage detection.

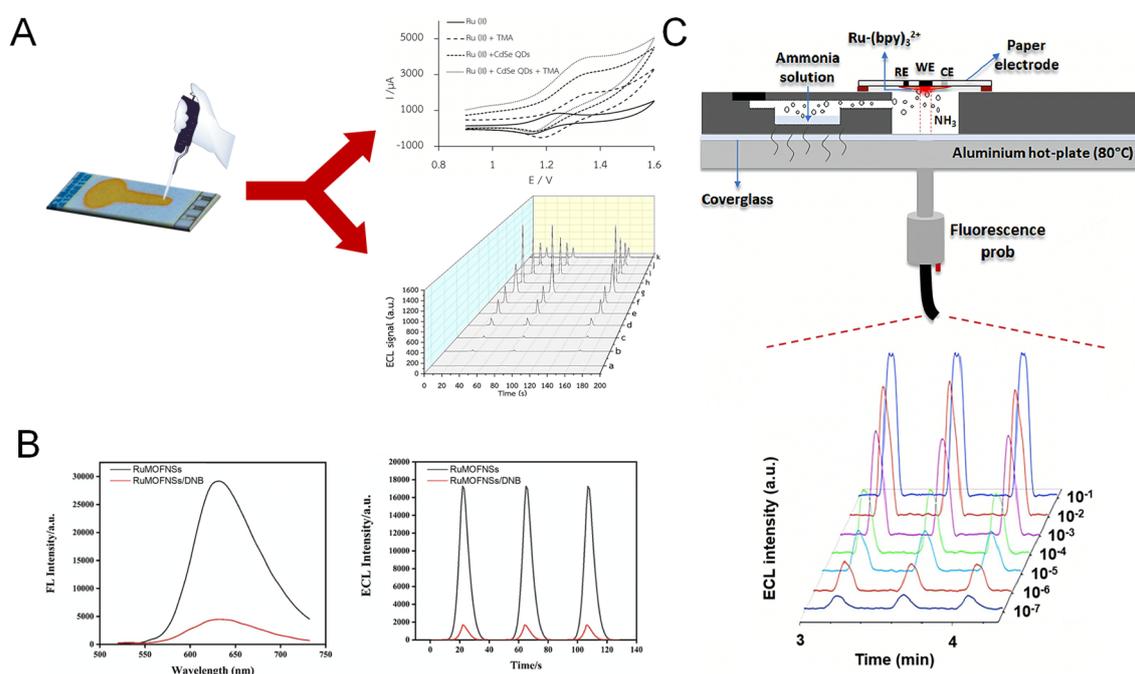


Figure 5. (A) Schematic of a paper-based electrochemiluminescence device for the estimation of trimethylamine. Reprinted with permission from Ref. [62]. Copyright 2022, Elsevier. (B) Schematic of FL and ECL profiles of RuMOFNSs. Reprinted with permission from Ref. [63]. Copyright 2025, Elsevier. (C) Schematic of the reaching of the ammonia gas onto the paper-based electrode surface by heating the ammonia solution in the reaction cell and the production of ECL signals. Reprinted with permission from Ref. [64]. Copyright 2022, Elsevier.

4. Conclusions and Perspectives

ECL-based sensing platforms have exhibited exceptional sensitivity and selectivity in detecting freshness-related analytes. In this review, we first provide an overview of the principles of ECL. We then summarize recent advances in ECL-based sensing platforms for the detection of biogenic amines, ATP and its degradation products, and volatile molecules, owing to their exceptional sensitivity and broad detection range. Despite these significant advances, the application of ECL in food freshness sensing has not yet been fully translated into practical and standardized sensing systems, and several challenges remain to be addressed.

(1) Enhancing stability and selectivity. The development of many ECL sensing platforms through multiple chemical or physical modification procedures is intended to enhance selectivity and signal output. However, these strategies often introduce challenges related to fabrication reproducibility and operational stability. Variations between sensor batches and challenges in maintaining long-term signal stability continue to limit practical application, particularly in on-site food analysis.

(2) Deepening mechanistic understanding of signal-amplification strategies. Significant advances have been made in developing enzymatic, nucleic acid, and nanomaterial-involved amplification strategies for enhancing

ECL responses. However, critical steps such as electron-transfer pathways, reaction kinetics, and the formation of transient species are often not clearly delineated. Advancing mechanistic insight into these fundamental processes will be essential for building amplification strategies that offer higher signal gains and more predictable analytical behavior.

(3) Developing multi-analyte sensing capability. Food matrices contain a wide range of chemically and biologically active compounds, which makes single-analyte detection inherently vulnerable to cross-reactivity and fluctuations in the surrounding environment. Because food freshness is determined by an interplay of biochemical transformations, microbial activity, and physicochemical changes, evaluating freshness through a single molecular marker often yields incomplete or potentially misleading information.

(4) Improving portability and field adaptability. The miniaturization of ECL sensing and its integration into portable platforms such as smartphones or point-of-care readers provides a practical pathway toward real-time, on-site freshness assessment. When combined with microfluidic sample manipulation, smartphone-based optical readout, and AI-assisted signal processing, portable ECL systems have the potential to deliver multidimensional and continuous freshness monitoring across the entire food supply chain.

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Conflicts of Interest

The authors declare no conflict of interest.

Use of AI and AI-Assisted Technologies

No AI tools were utilized for this paper.

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