

MXene@Au-functionalized Gel-Car-PVA hydrogel electrolyte for leak-free quasi-solid-state Zn-air fuel cells with uncertainty-aware trimodal sensing

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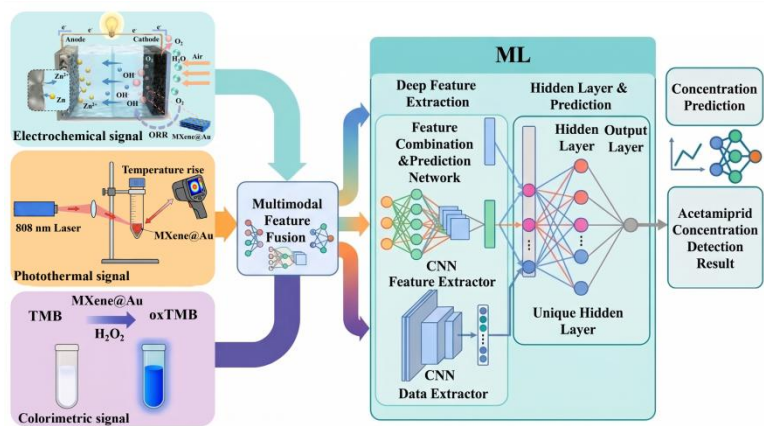
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This article reports a quasi-solid-state Zn-air fuel cell platform enabled by the synergistic design of materials innovation and explainable AI, which simultaneously addresses the two long-standing challenges of electrolyte leakage and AI model opacity, achieving reliable and accurate on-site detection of the pesticide acetamiprid.

MXene@Au-functionalized Gel-Car-PVA hydrogel electrolyte for leak-free quasi-solid-state Zn-air fuel cells with uncertainty-aware trimodal sensing

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ABSTRACT: The translation of self-powered electrochemical sensors from laboratory demonstrations to regulatory-compliant field deployment is impeded by two coupled barriers: (1) liquid electrolyte leakage and evaporation cause baseline drift under fluctuating conditions, and (2) black-box machine learning models lack uncertainty quantification and transparency for compliance testing. We report a co-designed quasi-solid-state Zn-air fuel cell platform that addresses both barriers through synergistic integration of materials engineering and algorithmic innovation, using acetamiprid (ACE) as a model pollutant. A physically cross-linked Gel-Car-PVA hydrogel electrolyte eliminates leakage and evaporation, suppressing signal attenuation to below 0.20% over 30 days across -20 to 25 °C, resolving the baseline instability that undermines field sensor credibility. Parallel to this, an uncertainty-calibrated one-dimensional convolutional neural network (1D-CNN) with Monte Carlo Dropout and SHapley Additive exPlanations (SHAP) delivers prediction intervals with quantified confidence mechanistic feature attribution, meeting regulatory transparency standards. This co-design enables a trimodal transduction strategy wherein aptamer-triggered release of MXene@Au nanohybrids generates electrochemical, photothermal, and colorimetric responses from a single assay, decoded by the interpretable AI framework. Benchmarking against liquid chromatography-tandem mass spectrometry (LC-MS/MS) in tomato, soil, and field water samples, the platform achieves quantitative agreement ($R^2 = 0.991$, slope = 0.83), confirming on-site screening reliability. This paradigm bridges the portability-accuracy gap in field-deployable trace pollutant surveillance, with implications for agricultural runoff and drinking water protection.

KEYWORDS: trimodal sensing; quasi-solid-state zinc-air fuel cell; gel-car-PVA hydrogel; uncertainty quantification; interpretable deep learning.

1 Introduction

The widespread application of neonicotinoid insecticides has been strongly linked to pollinator population declines [1], aquatic ecosystem impairment [2], and chronic dietary exposure risks in humans [3], driving an urgent need for high-frequency, large-scale monitoring across agricultural watersheds and drinking water sources [4]. Acetamiprid (ACE), one of the most widely used neonicotinoids with high water solubility (4 g/L) [5], exemplifies this class of polar, persistent trace organic contaminants and serves as the target analyte in this study [6]. While liquid chromatography-tandem mass spectrometry (LC-MS/MS) has been widely adopted for neonicotinoid quantification in aquatic and food matrices, the associated sample preparation steps often require laboratory conditions, organic solvents, and extended processing time, limiting its feasibility for high-frequency field screening [7-9]. To address this gap, aptamer (Apt)-based biosensors have emerged as a promising alternative for target-specific detection of neonicotinoids [10], with recent demonstrations including label-free detection of ACE in vegetable samples and DNzyme-based electrochemiluminescent quantification in food matrices

[11]. However, current implementations have not yet satisfied the requirements for regulatory acceptance, owing to two critical limitations: (1) conventional self-powered architectures with liquid electrolytes suffer from leakage, evaporation, and baseline drift under variable field conditions [12,13], and (2) machine learning models for signal decoding often operate as “black boxes” that lack uncertainty quantification and mechanistic interpretability, challenges that have been widely documented in both toxicological risk assessment and environmental modeling, and which limit their acceptance for compliance testing [14,15].

Zinc-air fuel cells (ZAFCs) have emerged as a promising self-powered platform for sensing applications owing to their high energy density and self-powered operation [16-18]. ZAFCs leverage abiotic oxygen reduction reaction (ORR) catalysts, enabling robust outdoor operation. Recent efforts have replaced liquid electrolytes with quasi-solid-state hydrogels to mitigate leakage, yet most hydrogel-based systems still suffer from mechanical instability and insufficient ionic conductivity under fluctuating temperature, humidity, or complex sample matrices [19-21]. To date, however, no hydrogel electrolyte has been reported that simultaneously supports multimodal transduction

(electrochemical, photothermal, colorimetric), maintains $<0.2\%$ signal drift over month-long deployment, and operates across -20 to $25\text{ }^{\circ}\text{C}$.

Addressing the first challenge requires moving beyond conventional liquid electrolytes toward quasi-solid-state architectures that withstand mechanical and thermal fluctuations in agricultural settings. Yet stabilizing the electrolyte alone is insufficient for sensitive detection. Hybrid nanomaterials combining MXene with metallic catalysts have been explored to amplify electrochemical signals through cascading catalytic strategies [22-25]. Transitioning from single-mode to multimodal sensing, integrating electrochemical, photothermal, and colorimetric outputs, represents a valuable advancement for regulatory screening. Such redundancy provides a robust strategy to discriminate true analyte signals from background noise in complex matrices, where a single readout is easily confounded by matrix effects.

The second unmet need is the lack of transparent, uncertainty-aware data analytics frameworks capable of decoding heterogeneous multimodal signals, a challenge that cannot be resolved by conventional calibration approaches either. Traditional linear calibration methods, including ordinary least squares and partial least squares regression, fail to capture the complex physicochemical interplay between the sensor interface and diverse environmental matrices [26-28]. While deep learning models including convolutional neural networks (CNNs) have improved interpretation accuracy, nearly all existing implementations operate as “black boxes” that provide no prediction uncertainty or mechanistic insight into feature importance.[29,30] For environmental compliance testing, this opacity is unacceptable, as regulators require calibrated confidence intervals near statutory thresholds and interpretable attribution to rule out matrix interference. Although recent studies have explored uncertainty quantification (UQ) and SHapley Additive exPlanations (SHAP) for laboratory analytical systems, none have adapted these tools to field-deployable multisensor platforms or validated them against reference methods in real-world agricultural samples [31,32]. Therefore, co-designing a robust sensing interface with a transparent, uncertainty-aware AI framework is essential to bridge laboratory demonstrations and field-deployable regulatory applications.

To address these dual barriers, we present an integrated quasi-solid-state ZAFs platform that couples a trimodal transduction interface with an uncertainty-aware deep learning framework for on-site pollutant detection. A physically crosslinked Gel-Car-PVA hydrogel eliminates leakage and resists degradation across -20 to $25\text{ }^{\circ}\text{C}$, while Apt-triggered release of MXene@Au nanohybrids simultaneously generates electrochemical, photothermal, and colorimetric readouts. This built-in signal redundancy suppresses false positives in complicated sample backgrounds, such as turbid surface water or clay-rich soils. To decode nonlinearly coupled multimodal responses, we integrate a one-dimensional CNN with Monte Carlo Dropout (MC-Dropout) for uncertainty quantification and SHAP for mechanistic interpretability. This end-to-end pipeline delivers concentration predictions with calibrated $\pm 2\sigma$ confidence intervals, equipping regulators with the transparency needed to assess measurement reliability near statutory

thresholds for ACE in drinking water and agricultural runoff. Beyond ACE, this integrated framework can be readily adapted for the detection of other neonicotinoids or analogous polar pesticides by substituting the corresponding aptamers.

2 Experimental

2.1 The reagents and experimental instruments

The reagents and experimental instruments are described in the Electronic Supplementary Material (ESM). All oligonucleotide sequences are listed in Table S1 in the ESM.

2.2 Preparation of MXene@Au composite

First, 3 mL of sodium citrate solution (2% (w/w)) was added to 350 mL of MXene dispersion (0.4 g/L; detailed synthesis procedures for MXene are provided in the ESM), followed by ultrasonication at room temperature for 30 min to achieve homogeneous dispersion. Subsequently, 540 μL of HAuCl_4 solution and 3.17 g of ascorbic acid (AA) were added to the mixture, and the reaction was allowed to proceed under continuous stirring at room temperature for 2 h to facilitate thorough in situ reduction. After the reaction, the resulting black precipitate was collected by centrifugation (11000 rpm, 10 min) and washed alternately with ultrapure water and absolute ethanol three times. Finally, the product, denoted as MXene@Au, was obtained after vacuum drying for 12 h.

2.3 Preparation of Gel-Car-PVA composite hydrogel

A one-pot strategy was employed to construct a physically crosslinked Gel-Car-PVA composite hydrogel network. First, a water/glycerol binary mixed solvent was prepared using 40 mL of deionized water and 10 mL of glycerol. Under constant magnetic stirring at $95\text{ }^{\circ}\text{C}$, 4 g of PVA, 1 g of carrageenan, and 1 g of gelatin powder were gradually added to the solvent, where pre-dispersion effectively prevented local agglomeration of the polymer chains. Under continuous heating and vigorous stirring (2-3 h), the components were fully dissolved, forming a homogeneous and transparent viscous precursor solution. After degassing by static standing at $95\text{ }^{\circ}\text{C}$ for 30 min, the precursor solution was promptly poured into a glass mold while hot. After naturally cooling to room temperature, three freeze-thaw cycles were applied to promote hydrogen bonding and physical crosslinking among the polymer chains. The resulting gel sample was sealed and stored at $4\text{ }^{\circ}\text{C}$ for subsequent measurements.

2.4 Preparation of F/M-MXene@Au

40 μL of the auxiliary strand M (1 μM) was mixed with 40 μL of MXene@Au (1.5 g/L) and incubated at $37\text{ }^{\circ}\text{C}$ for 1 h. Subsequently, 40 μL of the fuel strand F (1 μM), 10 μL of magnetic beads, and 10 μL of EDC/NHS were added, followed by another incubation at $37\text{ }^{\circ}\text{C}$ for 1 h.

2.5 Detection procedure

40 μL of Apt at a concentration of 1 μM was mixed with 40 μL of T solution in a centrifuge tube and incubated at $37\text{ }^{\circ}\text{C}$ for

1 h to form the Apt/T complex. Subsequently, 10 μL of the test solution was added to the complex and reacted at 37 $^{\circ}\text{C}$ for 1 h. After that, the F/M-MXene@Au composite was added, and the reaction continued at 37 $^{\circ}\text{C}$ for another 30 min. Upon completion of the reaction, a high-strength magnet was used to perform magnetic separation, allowing the magnetic beads to settle. The supernatant was then carefully collected to obtain the nucleic acid amplification product, M-MXene@Au.

2.6 Signal readout procedures

Upon completion of amplification, 20 μL of the product was sufficient for multimodal signal acquisition. The current signal was obtained by drop-casting the product onto a carbon cloth cathode and assembling a Zn-air battery, with the instantaneous current recorded after a brief equilibrium period. The colorimetric signal was measured by mixing the product with a 3,3',5,5'-Tetramethylbenzidine (TMB)/ H_2O_2 substrate solution and recording the blue intensity. For the photothermal signal, the product was diluted and exposed to an 808 nm laser for a short duration, and the temperature increase was monitored. The entire process was straightforward and required no complex separation or purification steps.

2.7 Sensor measurement

For sensor-based detection, all samples were used directly in aqueous form without any extraction, concentration, or cleanup steps. The filtrate obtained after 0.22 μm filtration was directly applied to the sensor for trimodal signal acquisition.

3 Results and discussion

3.1 Principles of the integrated sensing and AI fusion platform

To achieve high-sensitivity detection, the researchers designed an Apt-based strand displacement amplification strategy for tri-modal sensing of emerging contaminants, using ACE as a case example. As shown in Fig. 1(a), the target is first specifically recognized by the Apt, triggering the release of DNA strand T. Subsequently, the released strand T undergoes a strand displacement reaction with the M-F structure within the F/M-MXene@Au complex, leading to the detachment of the M component from the complex and yielding the nucleic acid amplification product M-MXene@Au. The MXene@Au within this amplification product can trigger triple response signals (electrochemical, photothermal, and colorimetric) under specific conditions. As illustrated in Fig. 1(b), when the MXene@Au-containing amplification product is drop-cast onto the surface of the air cathode, an instantaneous current signal can be measured using a digital multimeter. When it is mixed with a solution containing TMB and H_2O_2 , TMB is oxidized, producing a color change, and the resulting RGB (red-green-blue) Blue signal can be captured by a smartphone. Meanwhile, MXene@Au itself exhibits a good photothermal effect, generating a temperature change upon irradiation with an 808 nm laser, which can be recorded using a portable infrared thermal imager. On this basis, the self-powered sensor was further integrated with machine learning algorithms to develop an intelligent tri-modal detection platform. In this model, the current signal serves as the primary input feature, while the photothermal and colorimetric signals are embedded as key dimensions into the hidden layers of a neural network, where nonlinear weight allocation is accomplished through deep learning. Finally, using the real concentration of target as the output label, the network successfully achieves portable, on-site, and accurate detection.

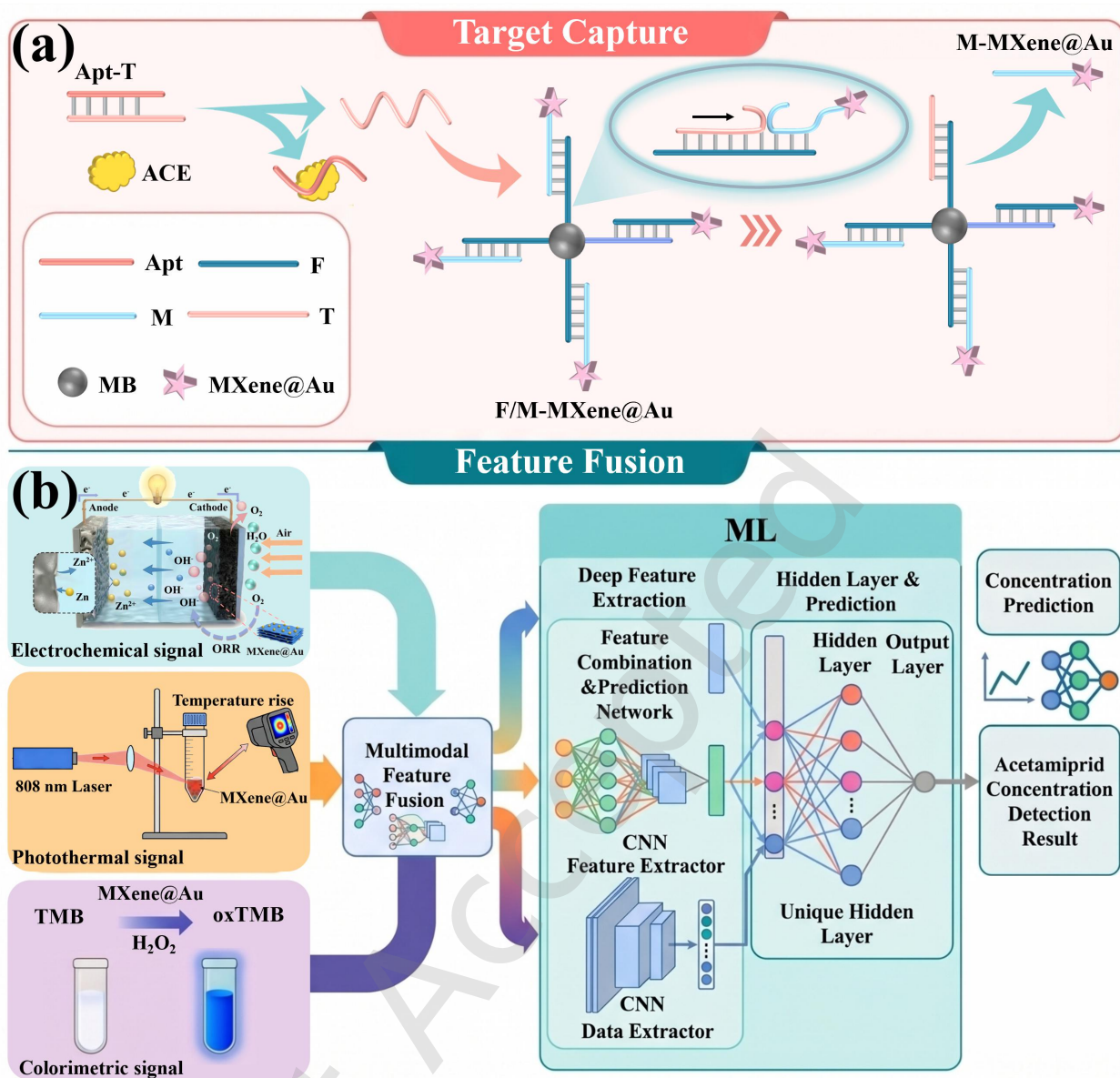


Figure 1 Schematic illustration of the uncertainty-aware multimodal self-powered sensing platform: (a) Workflow of the nucleic acid amplification-based recognition module, depicting the target-triggered strand displacement reaction and the liberation of MXene@Au nanohybrids for tri-modal signal transduction; (b) Architecture of the intelligent self-powered sensing platform, illustrating the synergistic integration of triple-mode signal outputs (current, photothermal, and colorimetric) with a machine learning (CNN)-driven data fusion engine for high-precision quantitative analysis.

3.2 Physicochemical characterization and device performance

Fig. 2(a) illustrates the stepwise fabrication of the Ti₃C₂ MXene support and its subsequent decoration with Au nanoparticles. Efficient selective etching of the Al atomic layers from the parent Ti₃AlC₂ MAX phase transforms the material into a well-defined, accordion-like lamellar morphology, as observed by scanning electron microscopy (SEM) (Fig. 2(b)). This structural transformation is further supported by X-ray diffraction (XRD) analysis (Fig. S1(a) in the ESM), where the characteristic (002) reflection shifts to $2\theta = 9.365^\circ$. Concurrently, the disappearance of the Al-related diffraction peak at 38.431° confirms the removal of the interstitial Al layers to below the XRD detection limit, consistent with successful exfoliation.

Leveraging the abundant surface reducing groups of the MXene matrix, an in-situ reduction strategy was employed to anchor Au nanoparticles. SEM (Fig. 2(c)) and transmission electron microscopy (TEM) (Fig. 2(d)) reveal a high-density, uniform distribution of Au nanoparticles across the MXene surface without apparent aggregation. High-resolution transmission electron microscopy (HRTEM) (Fig. 2(e)) reveals lattice fringes with spacings of 0.2219 nm and 0.2279 nm, corresponding to the (131) and (020) planes of metallic Au, respectively. The XRD pattern of MXene@Au (Fig. 2(f)) exhibits additional reflections at 37.95° , 44.18° , 64.37° , and 77.37° , indexed to the (111), (020), (022), and (131) planes of face-centered cubic Au (Ref. Code: 96-900-8464).

X-ray photoelectron spectroscopy (XPS) analysis was performed to investigate the surface chemical states of pristine MXene and MXene@Au. The Ti 2p spectra were deconvoluted into four spin-orbit doublets corresponding to Ti-C, Ti²⁺, Ti³⁺,

and Ti^{4+} (Fig. 2(g) and Table S5 in the ESM), which is consistent with previous reports [33]. Upon Au loading, the Ti 2p peaks exhibit a negative shift compared with pristine MXene, and the Au 4f spectrum (Fig. 2(h)) confirms the metallic state of Au (83.18/86.88 eV), indicating electron transfer from Au to MXene. This is further supported by the change in Ti 2p area ratios, where the high-valence Ti species ($\text{Ti}^{3+} + \text{Ti}^{4+}$) increase after Au loading.

SEM (Fig. 2(i)) reveals a highly interconnected, sponge-like 3D porous framework, which facilitates molecular diffusion and mass transport, attributes favorable for electrolyte infiltration and interfacial reactions in the sensing configuration. XPS and Fourier transform infrared spectroscopy (FTIR) spectroscopy were performed to characterize the chemical composition and bonding networks of the hydrogel. The XPS survey spectrum (Fig. 2(j)) shows C, N, and O as the predominant elements. The C 1s spectrum (Fig. 2(k)) is deconvoluted into three peaks at 284.03, 285.51, and 287.10 eV, assigned to C-C/C-H, C-O, and O-C=O, respectively. The N 1s spectrum (Fig. 2(l)) exhibits a single peak at 399.17 eV, corresponding to the C-N / amide groups from the gelatin backbone, confirming the successful incorporation of gelatin into the hydrogel network. The O 1s spectrum (Fig. 2(m)) shows a dominant peak at 531.65 eV, attributed to C-O/C=O species within the ternary network. FTIR (Fig. 2(n)) shows absorption bands corresponding to O-H/N-H (broad envelope), C=O, and C-O stretching vibrations, consistent with the hydrogen-bonding network inferred from XPS. These spectroscopic features are indicative of physically crosslinked domains that may contribute to the mechanical stability of the hydrogel matrix.

3.3 Multifunctional synergy of MXene@Au

The utility of MXene@Au as a trifunctional signal transducer was systematically evaluated across three modalities: electrocatalytic, photothermal, and colorimetric. The electrocatalytic performance was first examined by cyclic voltammetry (CV) and linear sweep voltammetry (LSV) in 0.5 M KOH. CV profiles under O_2 -saturated conditions (Fig. S2 in the ESM) show a distinct cathodic ORR peak that is absent under N_2 , confirming the oxygen-dependent nature of the response. LSV polarization curves (Fig. 3(a)) reveal a positive shift in onset potential for MXene@Au (0.8 V vs. RHE) relative to pristine MXene, indicating a lower thermodynamic barrier for ORR. Tafel analysis (Fig. 3(b)) gives a slope of $59.56 \text{ mV dec}^{-1}$

for MXene@Au, compared with $82.14 \text{ mV dec}^{-1}$ for pristine MXene, indicating faster ORR kinetics. Rotating disk electrode (RDE) measurements at varying rotation rates (Fig. S3 in the ESM) and the corresponding Koutecky-Levich plots (Fig. 3(c)) show good linearity, yielding an electron transfer number of $n \approx 2.0$, consistent with a dominant two-electron ORR pathway (see ESM for derivations).

To further corroborate the two-electron ORR pathway, rotating ring-disk electrode (RRDE) measurements were conducted to monitor the generation of peroxide intermediates. As shown in Fig. S4(a) in the ESM, the disk current (I_d) and ring current (I_r) were simultaneously recorded in O_2 -saturated 0.5 M KOH at 1600 rpm. The calculated electron transfer number is $n \approx 2.0$, and the H_2O_2 yield exceeds 85% (Fig. S4(b) in the ESM, see ESM for calculation details), confirming that the ORR on MXene@Au predominantly proceeds through a two-electron pathway with H_2O_2 as the primary product. This result is consistent with the Koutecký-Levich analysis, providing unambiguous experimental evidence for the proposed two-electron mechanism. Given that the sensor is designed for single-use field screening with a readout time of approximately 4 s, the H_2O_2 generated during this short measurement window does not appreciably compromise the instantaneous current output. This kinetic profile, lower overpotential and enhanced reduction current, is favorable for self-powered sensing applications.

The photothermal performance was evaluated under 808 nm irradiation. The steady-state temperature of the MXene@Au dispersion increased with power density from 0.5 to 1.5 W cm^{-2} (Fig. 3(d)). Cyclic on/off tests (1.0 W cm^{-2} , 600 s per cycle, Fig. 3(e)) showed negligible change in ΔT over three cycles, and the photothermal response remained stable after two months of ambient storage (Fig. 3(f)).

The peroxidase-like activity was assessed by UV-vis spectroscopy. In the presence of MXene@Au, H_2O_2 , and TMB, a characteristic absorption band at 650 nm emerged with a concomitant color change to deep blue (Fig. 3(g)). Control groups (without MXene@Au, without H_2O_2 , or without TMB) showed negligible absorbance changes, indicating that the nanozymatic oxidation requires the hybrid structure. To verify target-triggered signal generation, the ACE-responsive colorimetric response was examined. As shown in Fig. 3(h) and 3(i), introduction of ACE triggers nucleic acid amplification, which mediates TMB- H_2O_2 oxidation

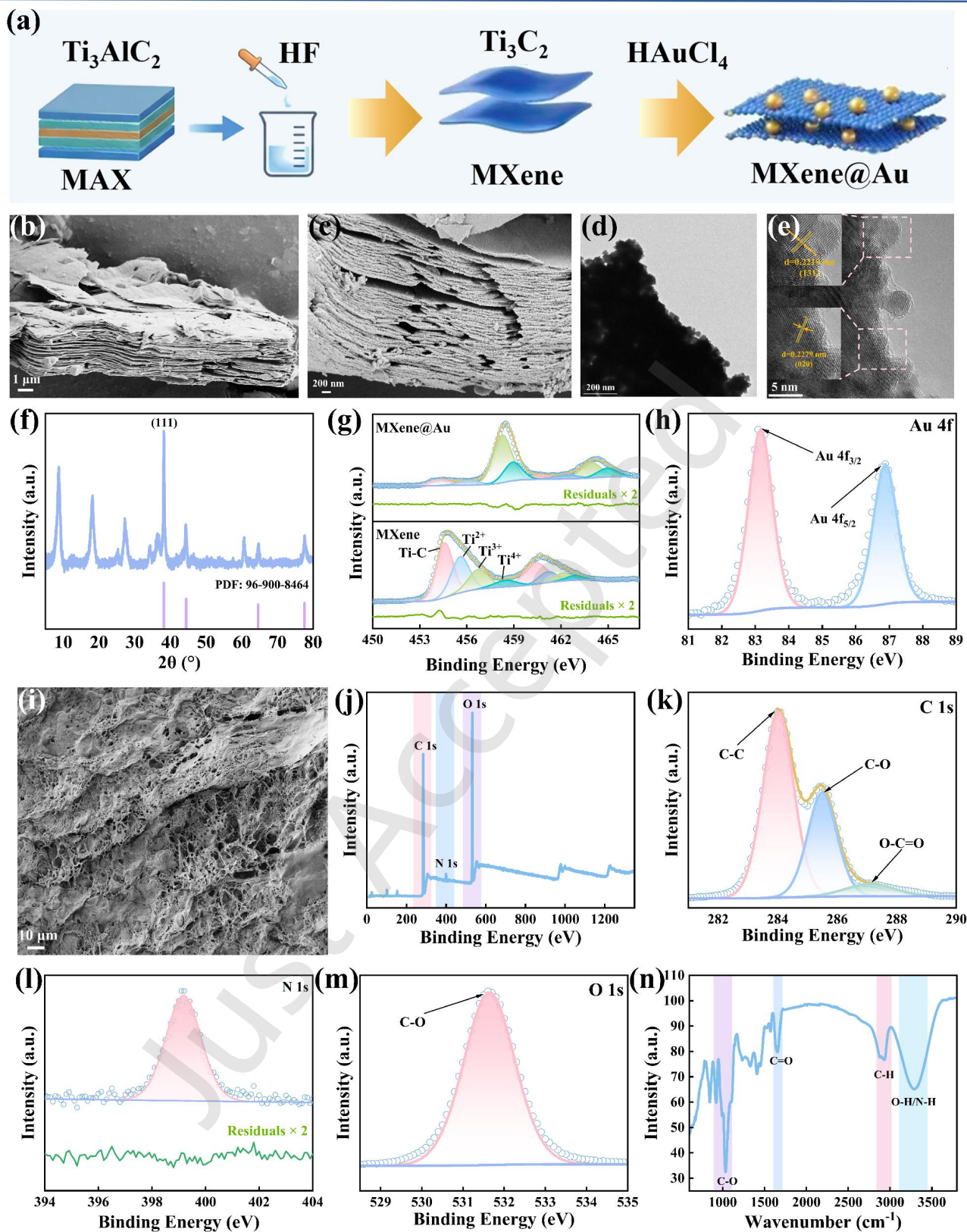


Figure 2 (a) Schematic illustration of the fabrication process for MXene and MXene@Au; (b) SEM images of MXene; (c) SEM and (d) TEM images of MXene@Au; (e) HRTEM image of MXene@Au; (f) XRD patterns of MXene and MXene@Au; XPS spectra: (g) Ti 2p of pristine MXene and MXene@Au, and (h) Au 4f of MXene@Au; (i) SEM image of the Gel-Car-PVA composite hydrogel; XPS spectra of the hydrogel: (j) survey scan, (k) C 1s, (l) N 1s, and (m) O 1s; (n) FTIR spectrum of the hydrogel.

and produces a concentration-dependent color change.

The integrated physicochemical properties of MXene@Au, ORR activity, photothermal conversion, and peroxidase-like nanozyme activity, were then harnessed in a trimodal sensing format (Fig. 3(j-l)). Upon target recognition, the Apt releases strand T, which initiates strand displacement on the F/M-MXene@Au track and liberates MXene@Au tags. These tags

simultaneously produce amplified cathodic current, temperature rise, and colorimetric readout from the same recognition event.

To evaluate the practical power output of the MXene@Au-based ZAFC for self-powered sensing, we assembled a quasi-solid-state Zn-air battery using the Gel-Car-PVA hydrogel electrolyte and measured its full-cell polarization and power density performance. As shown in Fig. S5 in the ESM, the assembled battery delivers an open-circuit voltage of 1.20 V and a maximum power density of 5.01 mW cm⁻² under ambient conditions. This power output is fully sufficient to drive low-power sensor modules and data transmission units, which typically require only tens to hundreds of microwatts. More importantly, the two-electron ORR pathway of MXene@Au (n

≈ 2.0) offers a distinct advantage for sensing applications: the in-situ generated H₂O₂ can directly participate in the colorimetric (TMB oxidation) readout, creating a seamless integration between the power-generation and signal-transduction functions. This eliminates the need for external power sources or additional redox mediators, significantly simplifying the device architecture and enhancing its portability for on-site field deployment. Furthermore, the quasi-solid-state hydrogel electrolyte effectively suppresses leakage and evaporation, ensuring stable long-term power output, a critical prerequisite for reliable agricultural environmental monitoring.

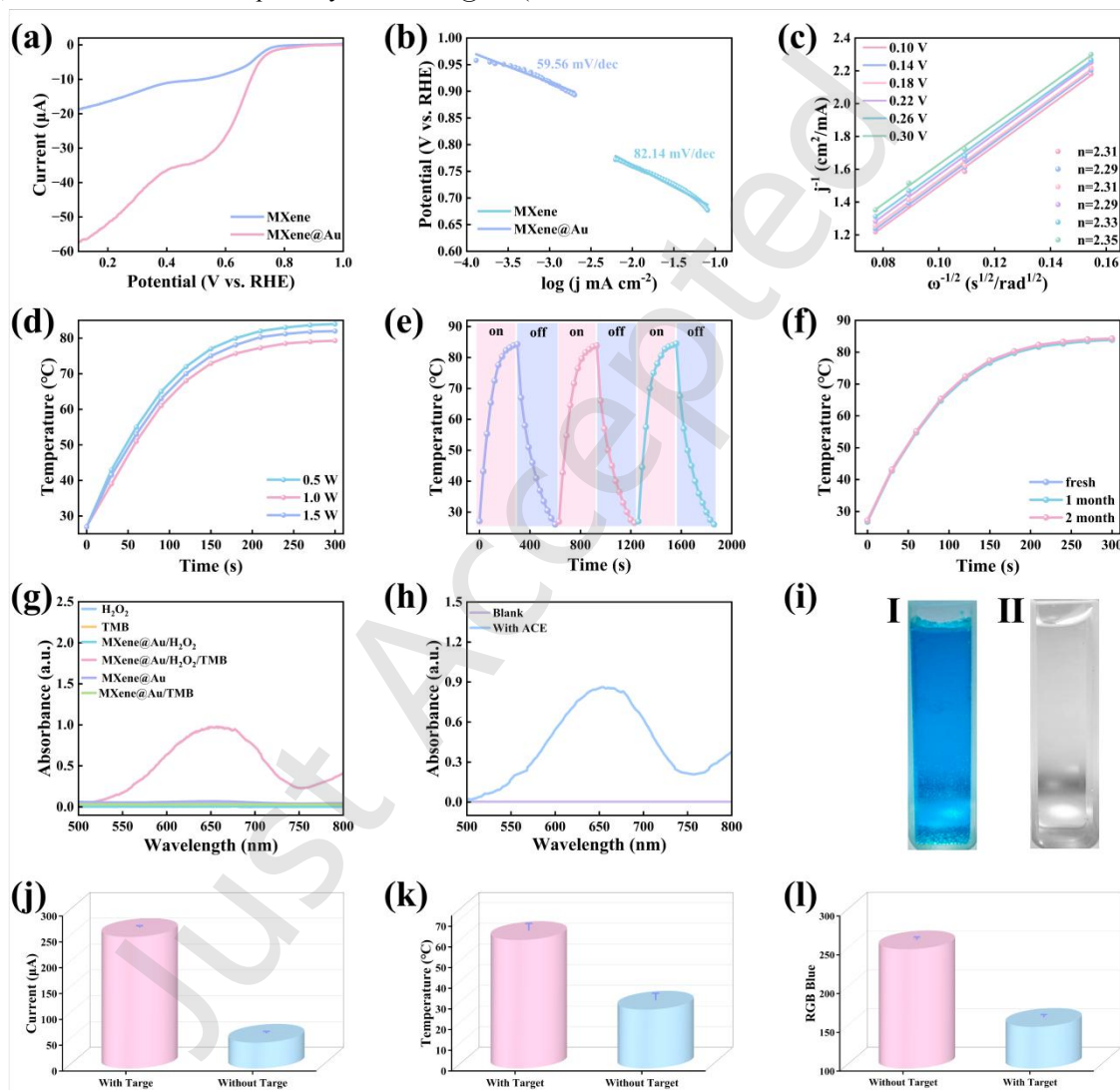


Figure 3 (a) LSV curve comparison of MXene and MXene@Au at 20 mV/s and 2000 rpm; (b) Tafel slope comparison of MXene and MXene@Au at 2000 rpm; (c) Electron transfer number of MXene@Au; (d) Photothermal heating curves of 1.5 g/L MXene@Au under different power irradiation (0.5, 1.0, 1.5 W); (e) Photothermal stability performance of MXene@Au under three cycles of 808 nm laser on/off switching; (f) Photothermal stability curve of MXene@Au over time; (g) Catalytic activity of MXene@Au; (h) Displacement of M-MXene@Au by T strand; (i) Photographs of the colorimetric reaction with ACE (I) and without ACE (II); (j) Current intensity in the presence or absence of target; (k) Photothermal intensity in the presence or absence of target; (l) Colorimetric intensity in the presence or absence of target.

3.4 Systematic evaluation of operational parameters for signal transduction

To achieve optimal detection performance of the sensor, a series of key experimental parameters were systematically

optimized. First, the effect of nucleic acid amplification time on the signal was investigated. As shown in Fig. 4(a), when the strand displacement reaction between DNA strand T and F/M-MXene@Au proceeds for 30 min, the current response of the system reaches its maximum value. Second, the working

concentration (Fig. 4(b)) and loading volume (Fig. 4(c)) of the material were optimized. The results indicate that at a concentration of 1.5 g/L and a loading volume of 40 μL , the released MXene@Au elicits the strongest current signal. Furthermore, KOH solution, employed as the electrolyte, possesses exceptionally high ionic conductivity, which significantly promotes the reaction kinetics at the sensing interface. The optimization results show that 0.5 M KOH

solution provides the optimal electrochemical reaction activity (Fig. 4(d)). Meanwhile, the optimal current output is achieved after immersing the hydrogel in KOH solution for 5 min (Fig. 4(e)). Finally, the thickness of the hydrogel is investigated, and it is experimentally demonstrated that a hydrogel film with a thickness of 1 mm yields the most favorable electrochemical signal response (Fig. 4(f)).

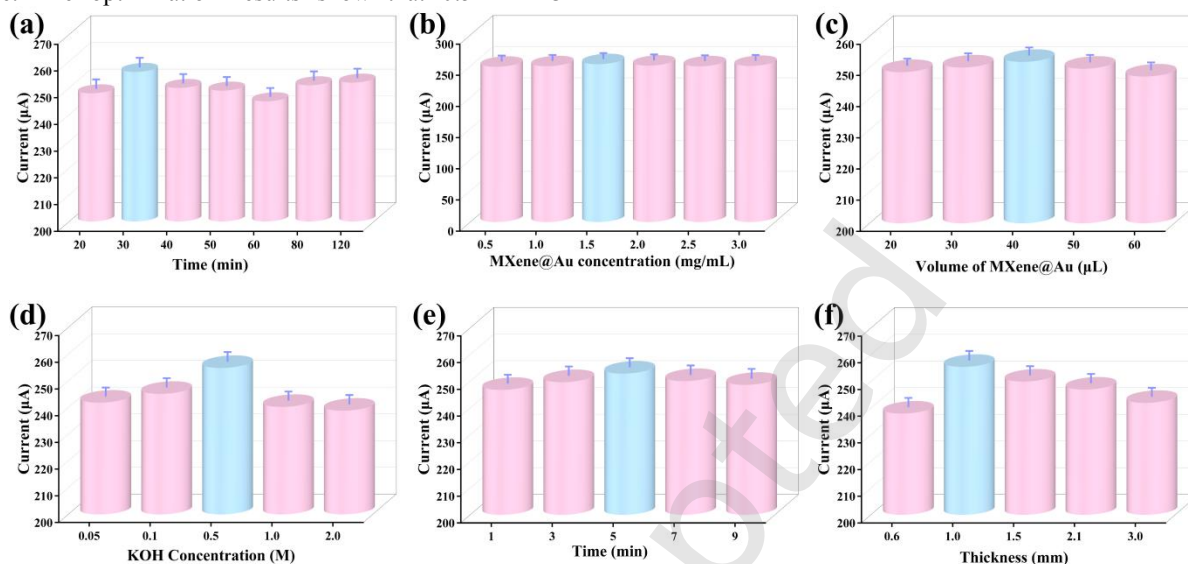


Figure 4 Optimization of key parameters for the self-powered sensing platform: effect of nucleic acid amplification time (a), MXene@Au concentration (b), MXene@Au loading volume (c), KOH electrolyte concentration (d), hydrogel soaking time (e), hydrogel thickness (f).

3.5 Assessing sensor reliability and real-world applicability

To validate the practical applicability of the trimodal platform, its operational stability, reproducibility, and selectivity were systematically examined across the three transduction channels.

Electrochemical mode. The storage stability of the hydrogel electrolyte was examined under thermal storage at -20, 4, and 25 $^{\circ}\text{C}$ over 30 days, with fresh aliquots sampled at intervals for single measurements. The self-powered current output toward 10^{-8} g/L ACE over 30 days (Fig. 5(a)) showed signal attenuation below 0.20% across all conditions. Sensor-to-sensor reproducibility, evaluated using independently fabricated batches (Fig. 5(b)), gave RSD values of 0.49-0.57%. Cross-reactivity tests with coexisting interferents, deoxynivalenol (DON), zearalenone (ZEN), aflatoxin B1 (AFB1), patulin (PAT), and Atrazine (ATZ) showed no significant perturbation of the electrochemical signal (Fig. 5(c)).

Photothermal mode. The stability of the nucleic acid amplification components, which govern the photothermal signal output, was assessed under the same storage conditions. After 30 days, the photothermal response for 10^{-8} g/L ACE (Fig. 5(d)) showed attenuation below 0.33%. Batch-to-batch reproducibility (Fig. 5(e)) yielded RSD values of 1.23-1.55%. In the presence of a mixed interferent solution (DON, ZEN, AFB1, PAT, and ATZ), the photothermal signal remained selective for ACE (Fig. 5(f)).

Colorimetric mode. The colorimetric channel was validated under identical storage and interference conditions. The colorimetric response (Fig. 5(g)) remained stable over 30 days with attenuation below 0.16%. Inter-batch reproducibility (Fig. 5(h)) gave RSD values of 0.55-0.70%. Neither visual nor spectrophotometric signals showed significant change upon exposure to the same panel of interferents (DON, ZEN, AFB1, PAT, and ATZ) (Fig. 5(i)), confirming the selectivity of the trimodal platform in multi-dimensional analysis.

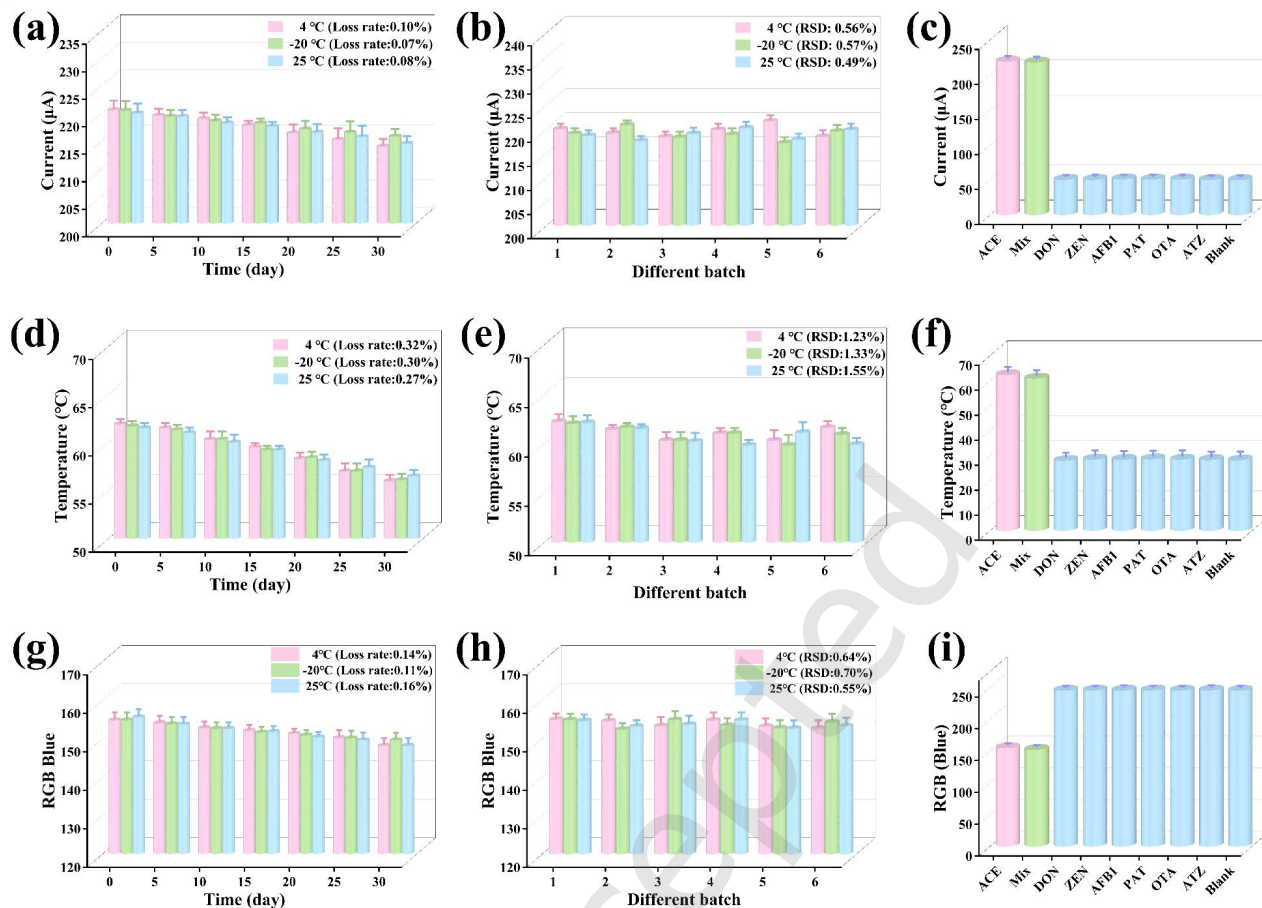


Figure 5 Sensor robustness, selectivity, and quality assurance. Electrochemical mode: (a) Stability, (b) reproducibility, and (c) selectivity. Photothermal mode: (d) Stability, (e) reproducibility, and (f) selectivity. Colorimetric mode: (g) Stability, (h) reproducibility, and (i) selectivity.

3.6 Sensing mechanism elucidation and interpretable model decisions

To elucidate the response characteristics of the trimodal sensor, exploratory data analysis was performed on the three signal features, electrochemical current, photothermal temperature, and colorimetric RGB Blue intensity. The violin plot (Fig. 6(a)) shows a high signal-to-noise ratio and measurement stability across repeated sampling. Principal component analysis (PCA) (Fig. 6(b)) shows that the first three principal components capture the majority of feature variance, confirming that the three signal modalities are not highly redundant and that the feature space is of low intrinsic dimensionality. The nonlinear trajectory in the three-dimensional feature space (Fig. 6(c)) further suggests that a linear mapping would be insufficient to capture the concentration-dependent signal evolution, motivating the use of a nonlinear model. The joint probability distribution (Fig. 6(d)) reveals enhanced sensitivity of the RGB Blue intensity at specific concentration ranges, consistent with the chromogenic kinetics of the TMB-based assay. For model development, the dataset was stratified by concentration and randomly divided into training (70%) and test (30%) sets. Data augmentation was performed only on the training set. Five-fold cross-validation was used within the training set for model selection, and the final model was assessed on the test set. The same splits (seed = 42) were used for all benchmark models.

Given the nonlinear nature of the feature manifold, a 1D-CNN was employed for signal decoding. The training dynamics (Fig. 6(e)) show convergence to an error floor on the order of 10^{-2} without oscillations under a cosine annealing scheduler, indicating stable learning. The parity plot (Fig. 6(g)) shows that predicted concentrations cluster along the $y = x$ diagonal across the entire dynamic range. Prediction residuals follow a Gaussian distribution (Fig. 6(f)), as confirmed by the Q-Q plot (Fig. 6(l)), and the residual scatter shows constant variance with no concentration-dependent trends (Fig. 6(m)), together suggesting that the model does not exhibit significant systematic bias over the tested concentration range. Benchmarking against Random Forest and XGBoost under five-fold cross-validation (Fig. 6(h)) shows that the 1D-CNN achieves the lowest root-mean-square error (RMSE), demonstrating superior adaptability to the trimodal sensing data.

To interpret the model, SHAP analysis was applied. The global SHAP summary (Fig. 6(j)) identifies the RGB Blue channel as the most influential contributor to concentration prediction, a finding robustly cross-validated by permutation importance (Fig. 6(i)). Local dependence analysis (Fig. 6(n)) reveals pairwise interactions between RGB Blue intensity and electrochemical current at high concentration levels, providing insight into global feature behavior. For individual samples, the waterfall plot (Fig. 6(k)) breaks down the decision trajectory, illustrating how feature contributions aggregate to a specific prediction. The 2D decision boundary (Fig. 6(o)) further

illustrates how the combination of RGB Blue and current signals collectively defines the prediction regions.

Beyond feature interpretation, model reliability was assessed via uncertainty quantification. MC-Dropout was performed for Bayesian approximate inference, mapping a $\pm 2\sigma$ confidence interval for each analytical node (Fig. 6(p), MC-Dropout implementation details in the ESM). The positive correlation between predicted standard deviation and actual

error (Fig. 6(r)) confirms the model's capacity to capture its own predictive uncertainty. Based on this calibrated uncertainty, the upper confidence bound (UCB) acquisition function was further applied to visualize the parameter space (amplification time and material loading) and identify regions where high confidence and high accuracy coexist (Fig. 6(q-s)). This analysis provides a proof-of-concept demonstration of how uncertainty-guided exploration can inform subsequent experimental design.

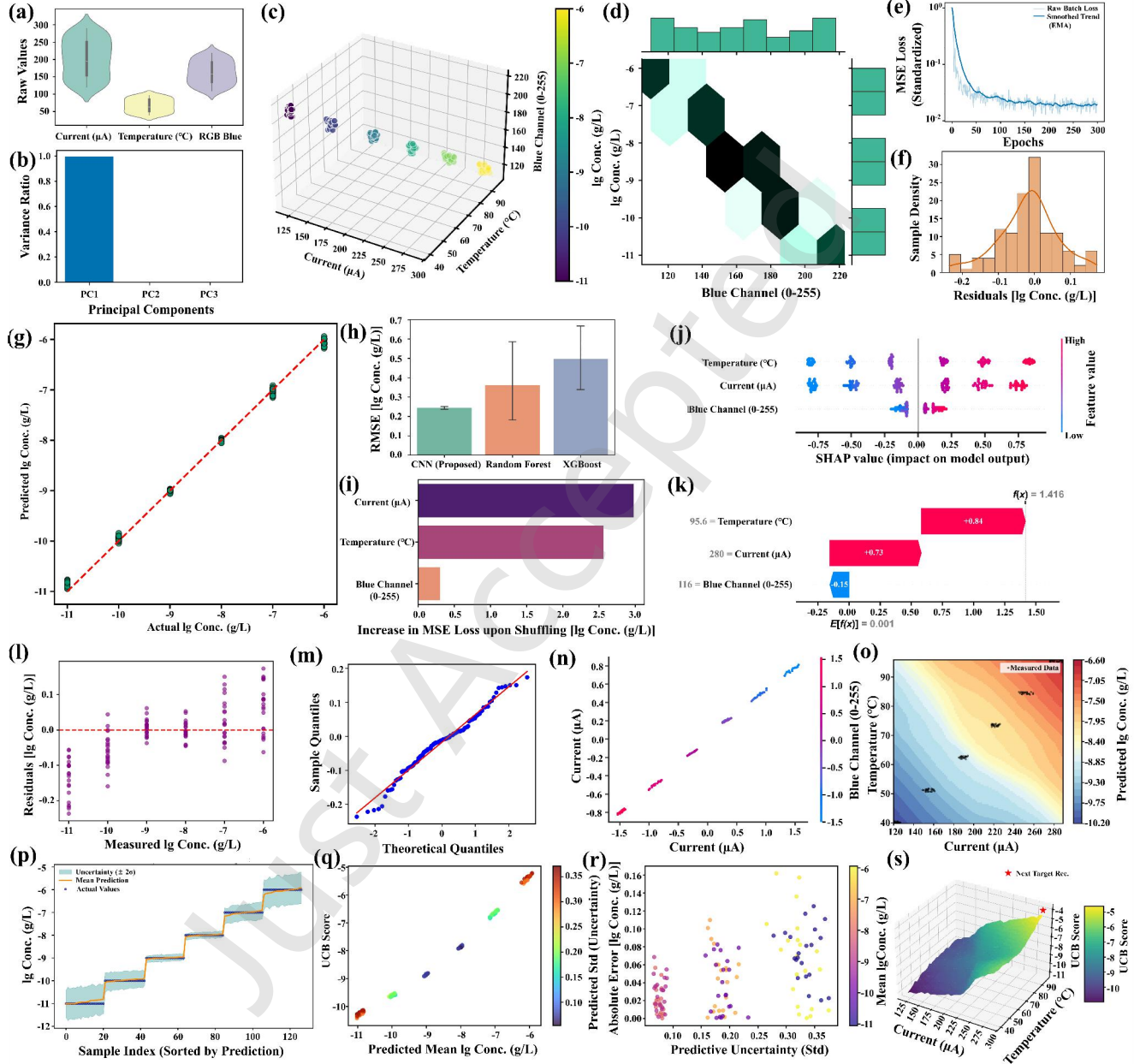


Figure 6 Machine learning-assisted signal decoding and model interpretability: (a) Violin plot of the three input features (current, temperature, RGB Blue); (b) Variance explained by the first three principal components; (c) 3D scatter plot of target concentrations in the feature space; (d) Joint distribution of RGB Blue intensity versus logarithmic concentration; (e) Training loss convergence of the 1D-CNN model over epochs (raw batch loss and EMA-smoothed trend); (f) Histogram of prediction residuals with fitted normal distribution curve; (g) Predicted versus actual concentrations (red dashed line: $y = x$); (h) RMSE comparison between 1D-CNN, Random Forest, and XGBoost under cross-validation; (i) Permutation feature importance; (j) Global SHAP feature importance and directional impact; (k) Waterfall plot of feature contributions for a representative high-concentration sample; (l) Q-Q plot of residuals for normality testing; (m) Residual scatter versus actual concentration; (n) SHAP dependence plot for the current feature; (o) 2D predicted response surface of RGB Blue versus temperature (RGB Blue fixed at mean); (p) Sample-wise predictions with $\pm 2\sigma$ confidence intervals via MC-Dropout; (q) 3D scatter plot of prediction mean, uncertainty (standard deviation), and UCB score; (r) Model-predicted uncertainty versus actual absolute prediction error; (s) 3D UCB score response surface in the parameter space.

3.7 Quantitative prediction and accuracy Validation in real samples

To validate the quantitative fidelity of the trimodal platform, we benchmarked its predictions against LC-MS/MS (GB/T 23584-2009) as the reference method. A calibration curve was first established using ACE standards (Figs. S6 and S7 in the ESM). The resulting linear regression was then applied to quantify ACE concentrations in seven authentic samples: tomatoes (samples 1, 2), adjacent soil (samples 3, 4), and field waterlogging (samples 5, 6), with a commercial ACE quality control sample in cabbage homogenate (sample 7, certified range 0.23-0.33 mg/kg) included as a quality control. The predicted ACE concentrations generated by the multimodal fusion framework are illustrated in Fig. 7(a); the quality control sample was determined to be 0.245 mg/kg by the sensor and 0.222 mg/kg by LC-MS/MS, both falling within the certified range.

To account for the discrepancy between single-channel and trimodal fusion predictions, we compare their conditional predictive distributions: $P(y|\text{channel})$ for any single modality (RGB Blue, current I, or temperature T), and $P(y|\text{RGB}, I, T)$ for the trimodal fusion model. As illustrated in Fig. 7(b), all three single-channel predictions exhibit notable deviations from the fusion results, though the magnitude and direction of deviation vary across channels. This follows from the fact that no single channel is sufficient to determine y ; $P(y|\text{channel})$ therefore represents only a partial view of the predictive distribution. Only when all three channels are jointly considered does the conditioning set become sufficiently informative to approximate the underlying relationship [34]. This divergence can be expressed as:

$$y_{\text{fusion}} = y_{\text{channel}} + \beta_{\text{channel}} \Delta x_{\text{channel}} + \varepsilon$$

where $\Delta x_{\text{channel}}$ captures the information contained in the other two channels that is orthogonal to the selected single channel [35]. For instance, when RGB Blue is the baseline, $\Delta x_{\text{RGB}} = [I - E(I|\text{RGB}), T - E(T|\text{RGB})]$; when I is the baseline, $\Delta x_I = [\text{RGB} -$

$E(\text{RGB}|I), T - E(T|I)]$; and when T is the baseline, $\Delta x_T = [\text{RGB} - E(\text{RGB}|T), I - E(I|T)]$. In each case, β_{channel} is the corresponding weighting vector and ε is a zero-mean residual term. The same logic holds for any single channel: its prediction is incomplete, and the fusion model fills in the missing information from the other two modalities. In practice, current and temperature compensate for matrix effects that RGB Blue alone cannot capture, and the reverse is also true.

Cross-validation between the multimodal fusion predictions and LC-MS/MS reference values (Fig. 7(c), Fig. S8 in the ESM) shows strong linear correlation ($R^2 = 0.991$) with a slope of 0.83, with data points clustered along the $y = x$ line. Collectively, these results demonstrate that the trimodal platform achieves quantitative accuracy comparable to LC-MS/MS across tomato, soil, and field water samples, while the multimodal fusion strategy provides a clear advantage over single-channel readouts for matrix-resistant field analysis. Notably, the sensor-based detection requires no extraction, concentration, or organic solvent, unlike the LC-MS/MS procedure, substantially simplifying on-site operation (Table S3 in the ESM).

To further verify the selectivity of the trimodal platform, cross-reactivity tests were conducted with four representative coexisting contaminants: deoxynivalenol (DON), zearalenone (ZEN), aflatoxin B1 (AFB1), and patulin (PAT), which are commonly found alongside ACE in food and agricultural matrices. As shown in Fig. 5c, 5f, 5i, individual introduction of each interferent at an equimolar concentration level caused no significant perturbation to the electrochemical, photothermal, and colorimetric signals, respectively. Furthermore, under a mixed-interferent challenge (DON + ZEN + AFB1 + PAT), the photothermal channel remained selectively responsive to ACE (Fig. 5f). These results confirm that the aptamer-mediated recognition and the built-in signal redundancy collectively endow the platform with high selectivity against complex matrix interference, substantiating its practical applicability for on-site screening.

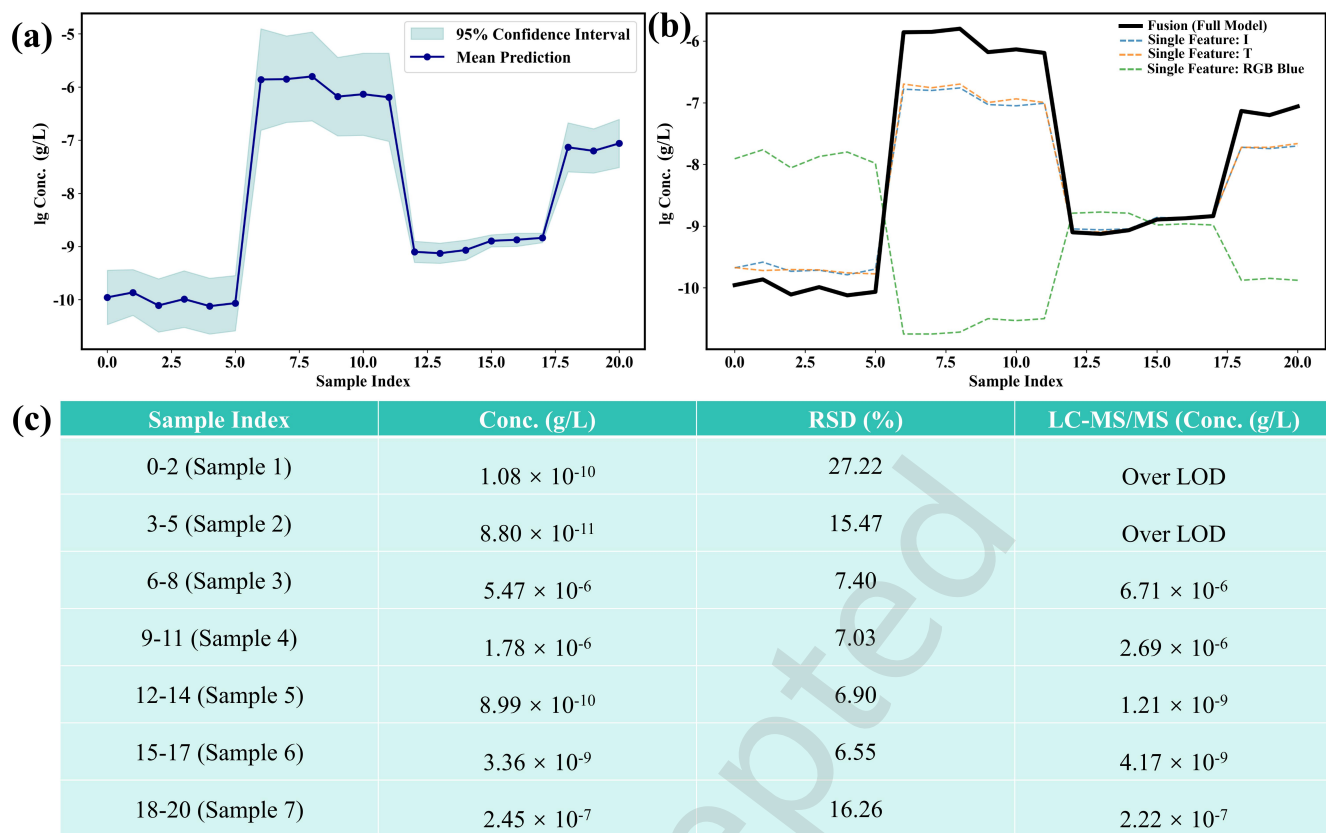


Fig. 7 (a) ACE concentrations predicted by the multimodal fusion framework for seven authentic samples (tomatoes, soil, field water, and a quality control); (b) Comparison of predictive performance between single features (I, T, RGB Blue) and multimodal feature fusion; (c) Correlation analysis between multimodal fusion predictions and reference values measured by LC-MS/MS.

4 Conclusions

In summary, we have developed a self-powered trimodal sensing platform that integrates a leak-free Gel-Car-PVA hydrogel electrolyte with MXene@Au nanohybrids as trifunctional signal transducers. The quasi-solid-state architecture effectively suppresses electrolyte leakage and ensures stable output across thermal extremes, while the MXene@Au tags concurrently generate electrochemical, photothermal, and colorimetric signals from a single recognition event, providing built-in redundancy against matrix interference. To decode these heterogeneous outputs, we employ a 1D-CNN augmented with MC-Dropout for uncertainty quantification and SHAP for interpretability. This framework delivers both concentration predictions and calibrated confidence intervals, enabling quantitative decision-making with rigorously quantified reliability.

Validation against LC-MS/MS in authentic tomato, soil, and field water samples establishes the quantitative reliability of the platform, with multimodal fusion consistently outperforming single-channel readouts. A commercial quality control sample in cabbage homogenate (certified range 0.23-0.33 mg/kg) was determined as 0.245 mg/kg by the sensor and 0.222 mg/kg by LC-MS/MS, both falling within the certified range, while correlation analysis yielded $R^2 = 0.991$ and a slope of 0.83. Critically, the interpretable AI component addresses a long-standing need in field sensor development: the ability to report not only a result, but also its associated confidence level, a

feature that distinguishes this platform from conventional “black-box” sensor designs.

We have demonstrated the platform for ACE as a model analyte; extending its applicability to other neonicotinoids or emerging contaminants would require corresponding Apt selection. While the hydrogel electrolyte shows stable performance under static storage, its mechanical durability under prolonged field handling warrants further investigation. Nevertheless, the integrated hardware-software architecture presents a viable pathway toward self-powered, AI-assisted environmental surveillance that bridges the gap between laboratory accuracy and field practicality.

Electronic Supplementary Material: Instrument, reagents, testing parameters, all oligonucleotide sequences required for the experiment, LC-MS/MS-related data, MXene@Au-related tests, and XRD data of MXene.

Data availability

All data supporting the findings of this study are available within the paper and its ESM.

Acknowledgements

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Declaration of competing interest

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

Author contribution statement

Kaili Wang: Investigation, Methodology, Writing-original draft. Chenchen Jin: Methodology, Formal analysis, Writing-review & editing. Guoxin Qin: Investigation. Yu Ya: Supervision. Lihui Mao: Supervision. Ning Li: Investigation, Formal analysis. Xuecai Tan: Conceptualization, Methodology, Writing-review & editing. Ke-Jing Huang: Conceptualization, Funding acquisition, Methodology, Project administration, Supervision, Writing-review & editing. All the authors have approved the final manuscript.

Informed consent

Not applicable.

Ethics statement

Not applicable.

Use of AI statement

None.

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Electronic Supplementary Material

MXene@Au-functionalized Gel-Car-PVA hydrogel electrolyte for leak-free quasi-solid-state Zn-air fuel cells with uncertainty-aware trimodal sensing

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Instrumentation and Materials

The morphological characteristics of the materials were characterized using a SUPRA 55 Sapphire scanning electron microscope (SEM) from Carl Zeiss AG, Germany, a FEI Talos 200S ultra-high-resolution field-emission transmission electron microscope (HRTEM) from Thermo Fisher Scientific (FEI), and Thermo Fisher Scientific ESCALAB Xi (XPS). X-ray diffraction (XRD) patterns were recorded on a Bruker D8 Advance A25 diffractometer (Bruker, Germany) with Cu K α radiation ($\lambda = 0.154$ nm). Perkin-Elmer Spectrum GX Fourier transform infrared spectrometer (FTIR) from PerkinElmer, USA. UV-Vis spectrophotometer (Evolution 220; Thermo Fisher Scientific, Waltham, MA, USA). Electrochemical measurements including cyclic voltammetry (CV) and linear sweep voltammetry (LSV) were performed using a PalmSens electrochemical workstation (Netherlands). A rotating disk electrode (RDE) system from Orignalys (France) was employed for hydrodynamic measurements. The instantaneous current output of the sensor was monitored using a UNI-T UT60BT Bluetooth digital multimeter (Uni-Trend Technology, China). An MDL-H-808-3W near-infrared laser device (Changchun New Industries Optoelectronics Technology, China) was used for photothermal excitation. According to GB/T 23584-2009, the LC-MS/MS separation was performed on a C18 column (50 mm $\times$ 2.1 mm, 1.7 μ m).

Acetamidrid (ACE, $\geq 97\%$), Zearalenone (ZEN, $\geq 97\%$), Deoxynivalenol (DON, $\geq 98\%$), magnetic beads, titanium aluminum carbide (Ti₃AlC₂, $\geq 98\%$), 40 wt% hydrofluoric acid (HF, ACS reagent grade), Gold chloride trihydrate (HAuCl₄, $\geq 99.9\%$ metals basis), Carrageenan, Adrenaline chloride tris(2-carboxyethyl)phosphine hydrochloride (TCEP, $\geq 98\%$), poly(vinyl alcohol) (PVA, type 1799, 98-99% hydrolysis), potassium hydroxide (KOH, AR), Hydrogen peroxide 30% aqueous solution H₂O₂ (GR), 3,3',5,5'-Tetramethylbenzidine (98%) were purchased from Aladdin Reagent Co., Ltd. (China). N-Hydroxysuccinimide (NHS) and 1-ethyl-3-(3-dimethylaminopropyl) carbodiimide hydrochloride (EDC) were obtained from Macklin Inc. (China). Acetamidrid quality control sample in vegetable matrix (cabbage homogenate) was purchased from Guangzhou Ceyan Technology Co., Ltd. The acetamidrid quality control sample in cabbage homogenate was prepared as a 1 mg/mL stock solution in ultrapure water. TE buffer and nuclease-free water were supplied by Solarbio Science & Technology Co., Ltd. (China). All oligonucleotides used in this study were synthesized by Sangon Biotech Co., Ltd. (Shanghai, China; sequences are listed in Table S1). BeyoMagTM Carboxyl Magnetic Beads (500 nm) were acquired from Beyotime Biotechnology (China). Zinc foil and carbon cloth were procured from online electrochemical material suppliers.

The aptamer sequences were adopted from a previously reported literature, with the specific sequence as follows: 1,2 5'- TGT AAT TTG TCT GCA GCG GTT CTT GAT CGC TGA CAC CAT ATT ATG AAG A -3'. He et al. obtained ACE-specific aptamer S18 ($K_d = 4.98$ μ M) via an immobilized ssDNA library-based SELEX strategy, which exhibited high selectivity against imidacloprid, nitenpyram, and chlorpyrifos.

Test Parameters

All electrochemical measurements including CV and LSV were carried out in a conventional three-electrode system. A reversible hydrogen electrode (RHE) and a graphite rod were used as the reference and counter electrodes, respectively. The scan rates for CV and LSV were set to 30 and 20 mV/s, respectively, all within specified potential windows. Data acquisition was controlled via the UNI-T Smart Test APP with a sampling interval of 0.3 s and an auto-stop time of 4 s. An MDL-H-808-3W near-infrared laser device was used for photothermal excitation.

XRD analysis was conducted using Cu K α radiation ($\lambda = 0.154$ nm) operated at 40 kV and 40 mA. The diffraction patterns were collected over a 2θ range of 5° to 80° with a step size of 0.02° and a scanning speed of 2° per minute. Data acquisition was performed using the UNI-T Smart Test APP, with a sampling interval set at 0.3 s and an automatic stop time of 4 s. X-ray diffraction (XRD) testing was conducted using Cu K α radiation, with a scanning angle ranging from 5° to 80°. The X-ray tube anode was made of copper, with a K α radiation wavelength of 1.5406 Å. The generator current and voltage were set at 40 mA and 40 kV, respectively. Image capture conditions: all images were captured inside a self-designed black box equipped with white LED lights to ensure uniform and consistent illumination. Application/software used: images were captured as raw files using the built-in camera application of a smartphone (Model: HUAWEI Mate 60) with fixed focus and exposure settings. The images were then transferred to a computer for subsequent analysis using a custom Python script. RGB extraction algorithm: for each test tube image, a fixed rectangular Region of Interest (ROI) was defined at the center of the reaction zone. The average RGB intensity values of the pixels within this ROI were extracted.

Table S1 Sequences of all oligonucleotides

Oligonucleotides	Sequences (5'-3')
Apt	TGT AAT TTG TCT GCA GCG GTT CTT GAT CGC TGA CAC CAT ATT ATG AAG A
T	TAG TCT CTA GAC AAA TTA CA
F	COOH-TGT AAT TTG TCT TGC TAA TCG TA
M	SH-CCA GAC TGG CAA ATT ACA

Synthesis of MXene

First, 2 g of Ti₃AlC₂ powder was slowly added to 50 mL of HF solution (40 wt%) under continuous stirring at 350 rpm. After the complete addition of the MAX phase, the polypropylene bottle containing the mixture was placed at 50 °C and stirred at 350 rpm for 48 h. After the reaction, the resulting precipitate was thoroughly washed and centrifuged with deionized water repeatedly until the pH of the supernatant reached above 6. Finally, the washed product was dried in a vacuum oven at 60 °C for 8 h to obtain multilayer MXene powder.

Koutecky-Levich equations:

$$j^{-1} = B^{-1} \omega^{-0.5} + j_k^{-1}$$

$$B = 0.62 n F C_0 D_0^{2/3} \nu^{-1/6} \omega^{-0.5}$$

In the equation, j (measured current density) and j_k (kinetic current density) are in mA cm^{-2} , ω is the rotation rate (rpm), and n is the electron transfer number. Standard values were employed for the constants: $F = 96500 \text{ C mol}^{-1}$, C_0 (O_2 in KOH) $= 1.2 \times 10^{-6} \text{ mol cm}^{-3}$, $D_0 = 1.9 \times 10^{-5} \text{ cm}^2 \text{ s}^{-1}$, and $\nu = 0.01 \text{ cm}^2 \text{ s}^{-1}$.

Calculation of electron transfer number (n) and H_2O_2 yield from RRDE measurements

The electron transfer number (n) and H_2O_2 yield were calculated from the RRDE data using the following equations:

$$n = 4 \times I_d / (I_d + I_r / N)$$

$$\text{H}_2\text{O}_2\% = 200 \times (I_r / N) / (I_d + I_r / N)$$

where I_d is the disk current, I_r is the ring current, and N is the collection efficiency of the Pt ring (0.37).

Sample preparation for LC-MS/MS reference measurement

For tomato samples, the edible portion was homogenized, and 5.0 g of homogenate was extracted with 10 mL of acetonitrile by vortexing (5 min) and ultrasonication (15 min), then centrifuged at 4000 rpm for 5 min. The supernatant was filtered through a $0.22 \mu\text{m}$ membrane prior to LC-MS/MS analysis.

For soil samples, air-dried and sieved (2 mm) soil (5.0 g) was extracted with 10 mL of acetonitrile-water (8:2, v/v) by vortexing (5 min) and ultrasonication (15 min), then centrifuged at 4000 rpm for 5 min. The supernatant was filtered through a $0.22 \mu\text{m}$ membrane prior to LC-MS/MS analysis.

For waterlogging samples, 100 mL of water was filtered through a $0.45 \mu\text{m}$ membrane to remove suspended solids and then filtered through a $0.22 \mu\text{m}$ membrane for LC-MS/MS analysis.

MC-Dropout implementation details

Uncertainty quantification was performed using Monte Carlo Dropout (MC-Dropout). For each test sample, 100 stochastic forward passes were conducted with the dropout layers kept active during inference. The dropout rates applied in the successive fully connected layers were 0.3, 0.2, and 0.1, respectively. The predictive mean and standard deviation (σ) were computed from the 100 predictions, and the $\pm 2\sigma$ interval was reported as the 95% confidence interval.

To assess the calibration of the prediction intervals, we computed the Pearson correlation coefficient between the predictive standard deviation (σ) and the absolute prediction error on the held-out test set. A positive correlation confirms that higher uncertainty corresponds to larger prediction errors, indicating that the model is well-calibrated.

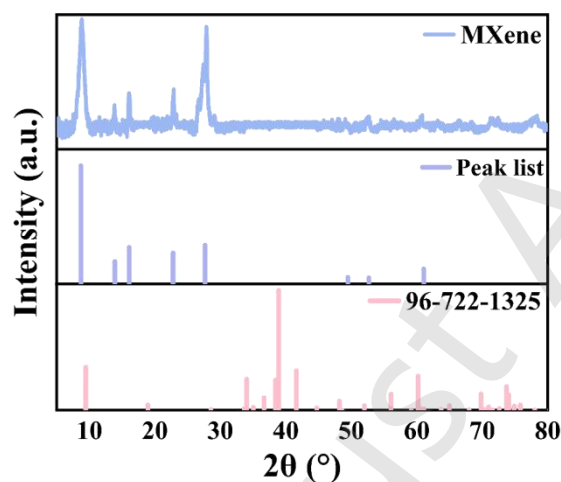


Figure S1 XRD data of MXene.

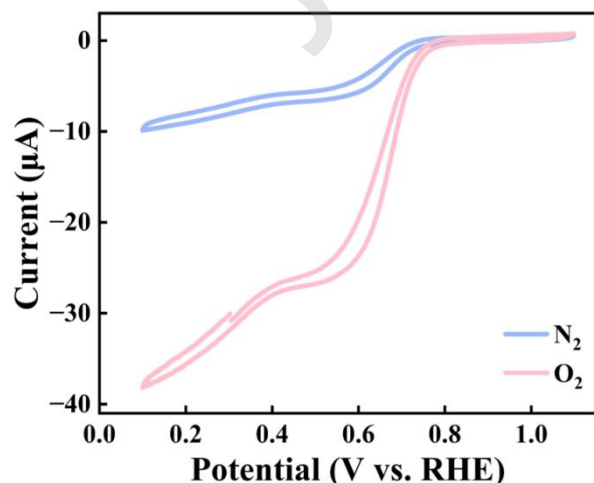


Figure S2 CV curves of MXene@Au under O₂-saturated and N₂-saturated conditions.

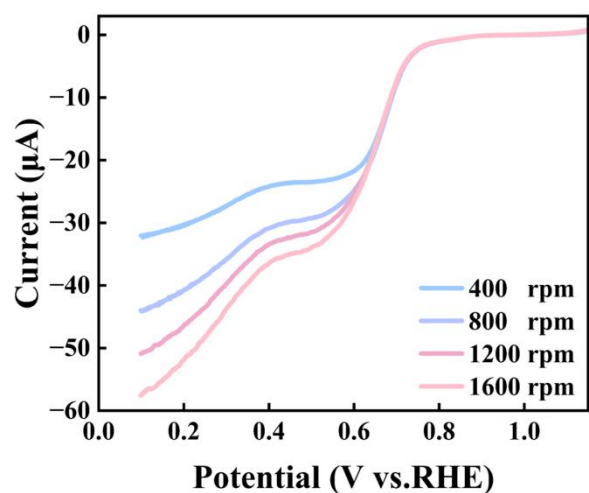


Figure S3 LSV curves of MXene@Au recorded at various rotation speeds in an oxygen-saturated environment.

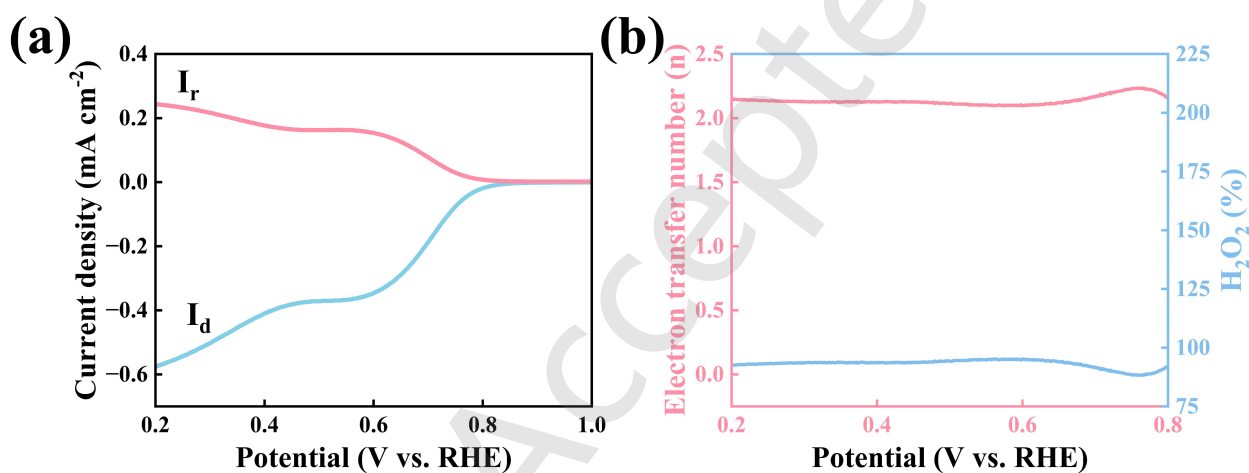


Figure S4 (a) RRDE voltammograms of MXene@Au in O₂-saturated 0.5 M KOH at 1600 rpm. (b) H₂O₂ yield and electron transfer number (n) versus potential.

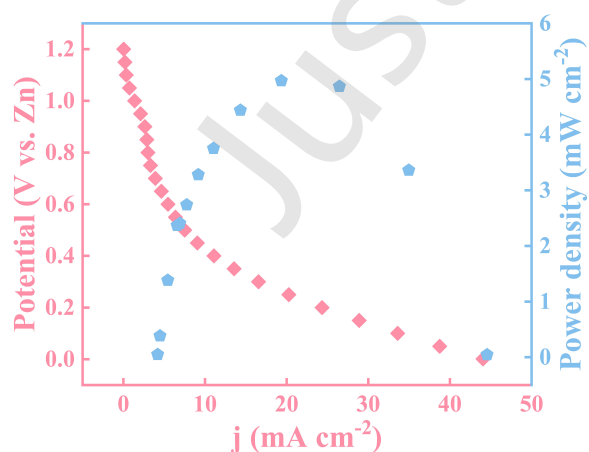


Figure S5 Polarization curve (left Y-axis, voltage versus current density) and corresponding power density curve (right Y-axis, power density versus current density) of the MXene@Au-based quasi-solid-state Zn-air battery.

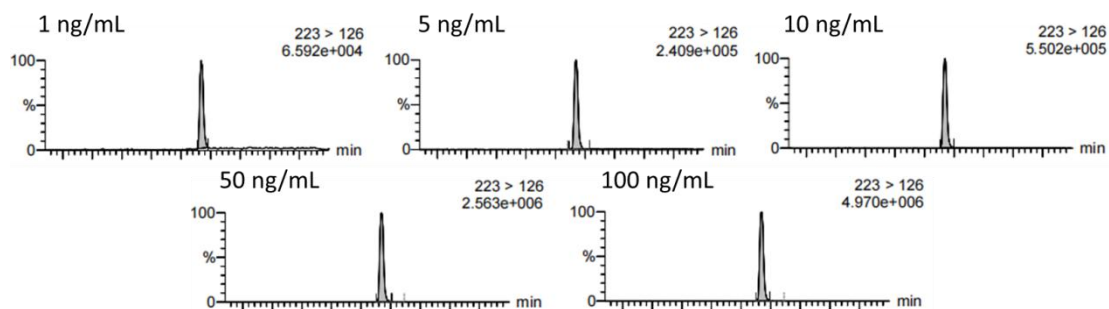


Figure S6 Liquid chromatograms of standard samples at different concentrations recorded via LC-MS/MS.

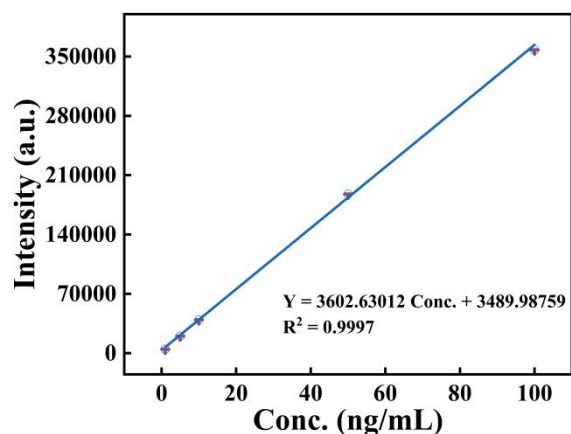


Figure S7 LC-MS/MS standard curve for acetamiprid.

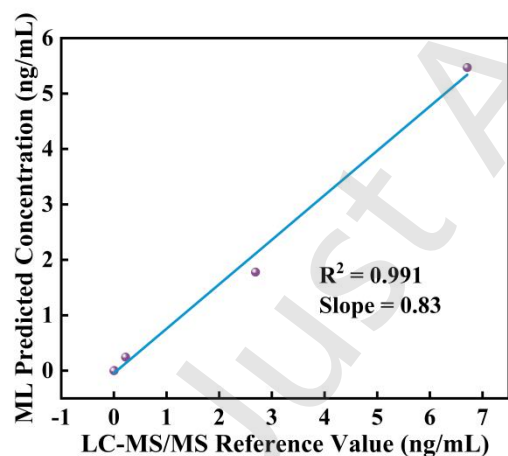


Figure S8 Correlation between ML-predicted acetamiprid concentrations (y-axis) and those determined by LC-MS/MS (x-axis).

Table S2 LC-MS/MS results of real sample analysis (n = 3)

Sample Number	Conc. (g/L)	RSD (%)
1	-4.55×10^{-7} (Over LOD)	3.45
2	-1.93×10^{-7} (Over LOD)	1.84
3	6.71×10^{-6}	0.17
4	2.69×10^{-6}	1.80
5	1.21×10^{-9}	0.83
6	4.17×10^{-9}	0.88
7	2.22×10^{-7}	1.92

Table S3 Comparison of analytical performance between the proposed trimodal sensor and LC-MS/MS (GB/T 23584-2009)

Feature	This work	LC-MS/MS
Sample preparation	Direct filtration (0.22 μ m)	Extraction, centrifugation, SPE cleanup

Organic solvent consumption	None	10–20 mL per sample (acetonitrile, methanol, etc.)
Measurement time per sample	4 s	10 min
On-site deployability	Yes (self-powered, portable)	No (laboratory-based)
Specialized training required	No	Yes

Table S4 Results of sensor spiking recovery experiment (n = 3)

Sample	Add (g/L)	Detect. (g/L)	Recovery (%)	RSD (%)
tomatoes	0	9.00×10^{-11}	-	4.21
	10^{-8}	1.03×10^{-8}	102.1	1.43
	10^{-9}	1.06×10^{-9}	97.0	4.79
	10^{-10}	1.90×10^{-10}	100.0	2.82

Table S5 XPS peak fitting parameters for the Ti 2p and N 1s spectra

Component	Peak	B.E. (eV)	FWHM (eV)	Area (CPS·eV)	Chi-squared value (χ^2)
Ti (MXene@Au)	Ti-C	454.33	1.65	5149.60	2.424
		460.66	1.64	3764.81	
	Ti ²⁺	456.12	1.80	4140.73	
		462.35	1.80	4169.29	
	Ti ³⁺	458.27	1.44	38390.44	
		463.92	1.60	13611.88	
	Ti ⁴⁺	458.93	1.40	14630.13	
		464.95	1.50	7440.81	
	Ti-C	454.58	1.28	35941.00	
		460.39	1.80	19597.62	
Ti 2p (MXene)	Ti ²⁺	455.58	1.42	26472.85	3.907
		461.16	1.42	7884.69	
	Ti ³⁺	456.74	1.58	16055.58	
		462.11	1.42	5809.28	
	Ti ⁴⁺	458.48	1.76	5970.51	
		462.87	1.34	3156.25	
Gel-Car-PVA Composite Hydrogel	C-N	399.17	1.61	9017.58	1.548

Reference

- [1] Shi, H.; Zhao, G.; Liu, M.; Fan, L.; Cao, T. Aptamer-based colorimetric sensing of acetamiprid in soil samples: Sensitivity, selectivity and mechanism. *J. Hazard. Mater.* **2013**, *260*, 754-761.
- [2] He, J.; Liu, Y.; Fan, M.; Liu, X. Isolation and Identification of the DNA Aptamer Target to Acetamiprid. *J. Agric. Food Chem.* **2011**, *59*(5), 1582-1586.