



Energy-saving hydrogen production coupled with PET-derived ethylene glycol oxidation: Reaction mechanisms, catalyst design, and reactor engineering

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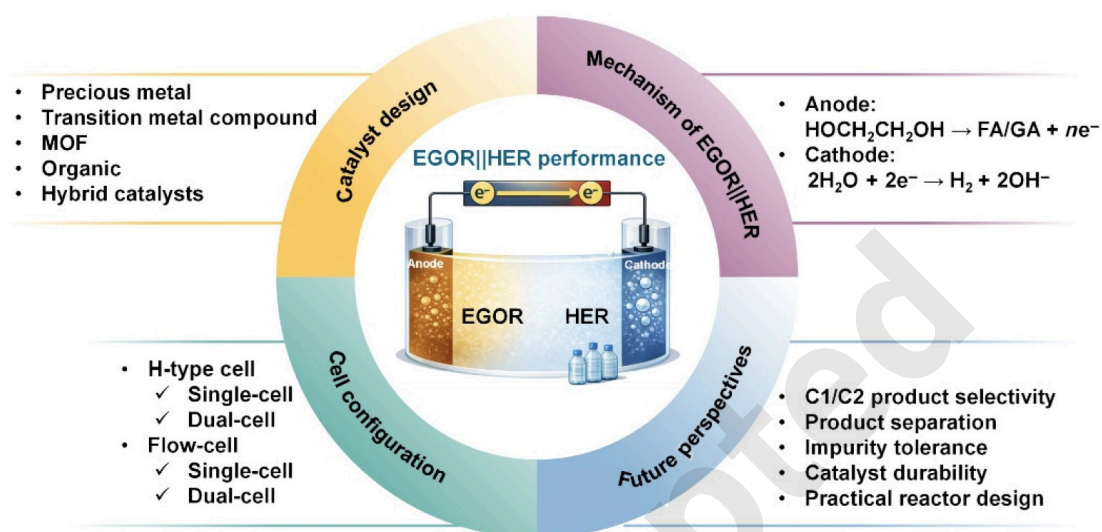
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ABSTRACT

The growing accumulation of poly(ethylene terephthalate) (PET) waste and the increasing demand for clean hydrogen call for integrated electrochemical strategies. Ethylene glycol oxidation reaction (EGOR), derived from PET hydrolysis, offers a thermodynamically favorable alternative to the sluggish oxygen evolution reaction (OER) and can be coupled with the hydrogen evolution reaction (HER) to simultaneously produce value-added chemicals and H₂. This review provides a system-level overview of EGOR||HER paired electrolysis, spanning reaction fundamentals, catalyst design, cell configuration, and future challenges. First, the competing EGOR pathways toward C1 products (such as formate/formic acid) and C2 products (such as glycolate/glycolic acid) are discussed. Key energetic descriptors, including cell voltage, overpotential, Faradaic efficiency, and energy efficiency, are then summarized to clarify the advantages of replacing OER with EGOR. Recent advances in catalysts, including precious metals, transition-metal compounds, metal–organic frameworks (MOFs), organic catalysts, and hybrid systems, are reviewed with attention to their bifunctional or compatible activity for EGOR and HER. Reactor design is further discussed by comparing single-cell and dual-cell H-type reactors with membrane-free and membrane-separated flow-cell reactors, emphasizing their different roles in mass transport, product separation, and scale-up feasibility. Finally, remaining challenges and future directions are proposed, including C1/C2 selectivity, downstream separation, impurity tolerance, catalyst durability, and practical reactor optimization. This review aims to guide the development of efficient EGOR-assisted hydrogen production systems for sustainable PET electro-upcycling.

KEYWORDS

PET electro-upcycling, ethylene glycol, paired electrolysis, bifunctional electrocatalysts, hydrogen

1 Introduction

The continuously increasing global demand for energy and the rapid accumulation of plastic waste have become two of the most pressing challenges for sustainable development [1–5]. Poly(ethylene terephthalate) (PET), one of the most extensively produced and consumed thermoplastics, is widely used in beverage bottles, food packaging, and synthetic fibers due to its excellent mechanical properties and chemical stability [6, 7]. However, these same characteristics also lead to its persistence in

the environment. Although PET is theoretically recyclable, the current recycling rate remains low, and a large fraction of post-consumer PET is landfilled, incinerated, or leaked into natural ecosystems [8].

Electrochemical technologies powered by renewable electricity provide a promising pathway for addressing both plastic waste and clean energy production [9]. PET can be hydrolyzed under relatively mild conditions to produce ethylene glycol (EG) and terephthalate [10], and the resulting EG can serve as an anodic

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substrate for electrochemical oxidation. Compared with the conventional oxygen evolution reaction (OER) in water electrolysis [11], the EG oxidation reaction (EGOR) is thermodynamically more favorable and kinetically less demanding [12, 13], because it avoids the energy-intensive O–O bond formation step associated with OER. As a result, replacing OER with EGOR can significantly reduce the anodic potential and overall cell voltage [14].

In addition, EGOR can be paired with multiple cathodic reactions [15], including the hydrogen evolution reaction (HER) [9], the two-electron oxygen reduction reaction to H_2O_2 (2e^- ORR to H_2O_2) [16], carbon dioxide reduction reaction (CO_2RR) [17], and nitrate reduction reaction ($\text{NO}_3^- \text{RR}$) [12], thereby enabling dual-functional electrolysis systems. Among these options, HER is the most energy-relevant counterpart owing to its well-established infrastructure, high reaction kinetics, and the direct production of carbon-free hydrogen fuel [18]. When coupled with HER at the cathode, this paired electrolysis strategy enables the simultaneous generation of value-added organic chemicals and green hydrogen with markedly enhanced energy efficiency [19–21]. This configuration not only reduces the electrical energy input for hydrogen production but also upgrades PET-derived feedstocks into high-value chemicals, thus establishing a synergistic platform that bridges plastic waste valorization with renewable energy storage [9].

Figure 1 shows a bibliometric analysis based on the Web of Science database using the keywords “PET + upcycling + electrochemical” and “PET + upcycling + electrochemical + hydrogen”, respectively. The rapidly increasing number of publications in Fig. 1(a) demonstrates the growing interest in

electrochemical PET upcycling as a sustainable alternative to conventional thermochemical processes. After introducing “hydrogen” as an additional keyword (Fig. 1(b)), although the total number of publications is lower, a much steeper growth rate is observed, indicating that coupling PET electro-upcycling with hydrogen production is an emerging research hotspot. The fast expansion, together with the still limited number of studies, highlights the timeliness and necessity of developing integrated systems for simultaneous plastic waste valorization and clean energy generation.

Despite the rapid growth of this field, current studies remain largely fragmented. Many reports focus on catalyst development or mechanistic investigations of EGOR alone [22–25], while the integration of reaction pathways, thermodynamic analysis, bifunctional catalysis, and reactor engineering is still insufficient [26, 27]. In most cases, electrocatalysts for EGOR and HER are developed separately. Because the anodic and cathodic reactions occur simultaneously in practical paired electrolysis, the catalysts for both half-reactions should be developed and evaluated together. In addition, reactor-level studies for EGOR||HER systems are still scarce. Rational reactor design is essential for improving mass transport and, more importantly, for enabling efficient product separation, which directly affects the overall process efficiency and techno-economic feasibility. A comprehensive and critical overview that connects these aspects is therefore urgently needed. Unlike previous catalyst-centered reviews, this work provides a system-level analysis of EGOR||HER paired electrolysis by linking reaction pathways, energetic parameters, bifunctional catalyst design, and reactor engineering. It further compares different catalyst and cell configurations in

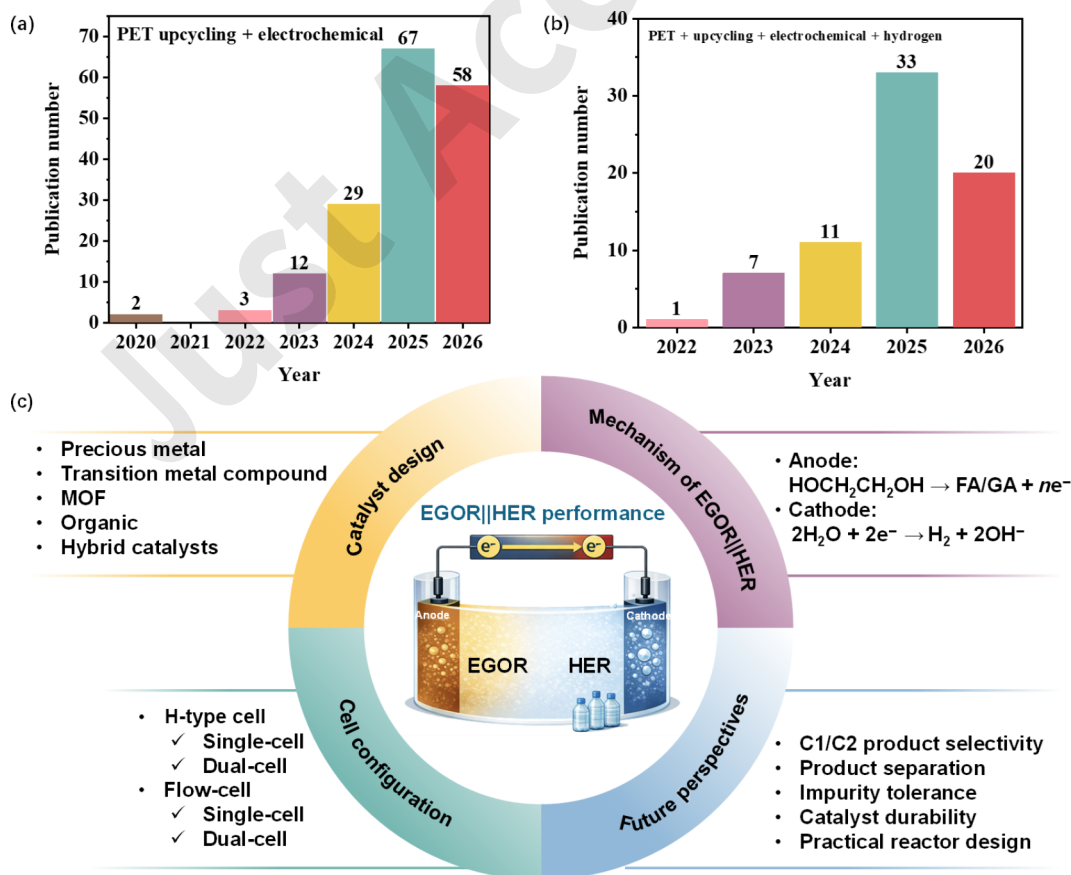


Figure 1 Number of publications retrieved from the Web of Science using the keywords (a) “PET + upcycling + electrochemical” and (b) “PET + upcycling + electrochemical + hydrogen” (search conducted on August 1, 2026). (c) Schematic overview of the EGOR||HER system discussed in this review.

terms of selectivity, energy efficiency, product separation, and scale-up potential. This integrated perspective provides practical guidance for advancing PET electro-upcycling toward efficient hydrogen production.

This review focuses on EGOR||HER coupled electrolysis as an emerging strategy for simultaneous ethylene glycol valorization and hydrogen production. As summarized in Fig. 1(c), EGOR||HER performance is placed at the center of this review, highlighting the core concept that anodic EGOR can replace the conventional OER and be coupled with cathodic HER to achieve value-added chemical production and energy-saving H_2 generation. Around this central theme, the review is organized into four major aspects. First, the reaction mechanism of EGOR||HER coupling is introduced, including anodic EG oxidation toward C1/C2 products and cathodic H_2 evolution. Second, different catalyst categories are summarized, including precious metals, transition-metal compounds, metal-organic frameworks (MOFs), organic catalysts, and hybrid catalysts. Third, various cell configurations are compared, including H-type cells, single-cell and dual-cell systems, and flow-cell reactors, emphasizing the importance of reactor design in mass transport, product separation, and practical operation. Finally, future research directions are discussed, including C1/C2 product selectivity, product separation, impurity tolerance of real PET waste, catalyst durability, and practical reactor design.

2 Fundamentals of EGOR coupled with HER

2.1 Anodic EGOR pathways

PET is first subjected to pretreatment and alkaline hydrolysis to release EG before electrochemical oxidation [28]. During hydrolysis, the ester bonds in the PET polymer chain are cleaved by OH^- , producing EG and terephthalate species, such as

disodium terephthalate or potassium terephthalate, depending on the alkaline electrolyte used. The resulting PET hydrolysate therefore contains EG as the main electroactive organic substrate, together with terephthalate salts, residual alkali, and possible impurities from real plastic waste. After filtration or basic purification to remove insoluble residues, the EG-containing hydrolysate can be directly introduced into the anodic compartment for EGOR. EG electro-oxidation mainly follows two pathways, producing C1 products represented by formic acid (FA)/formate and C2 products represented by glycolic acid (GA)/glycolate [29, 30], respectively (Fig. 2(a)).

At present, most reported EGOR systems still mainly target FA/formate, because deep oxidation and C–C bond cleavage are readily promoted on highly oxidizing transition-metal oxyhydroxide surfaces. In contrast, the C2 pathway preserves the C–C bond and is generally more favorable in terms of carbon utilization and energy consumption, but GA selectivity is more difficult to maintain under high-current operation. Economically, FA is a larger-volume bulk chemical but has a relatively low price, typically around USD 0.36–0.90 kg^{-1} , whereas GA is a higher-value specialty chemical, with bulk 70% GA listings generally around USD 1.9–2.2 kg^{-1} and small-volume/high-purity products being much more expensive. Therefore, although the C2 pathway offers intrinsic energy-saving and carbon-retention advantages, its practical economic benefit depends on whether high GA selectivity, low separation cost, and sufficient market demand can be achieved. By comparison, the C1 pathway may be more compatible with large-scale production, but its lower product value requires very high productivity, Faradaic efficiency (FE), and long-term stability to become economically attractive.

2.1.1 C1 pathway: EG to FA

The C1 pathway involves C–C bond cleavage of EG, leading to FA as the main product [31]. In a typical C1 pathway, EG is first

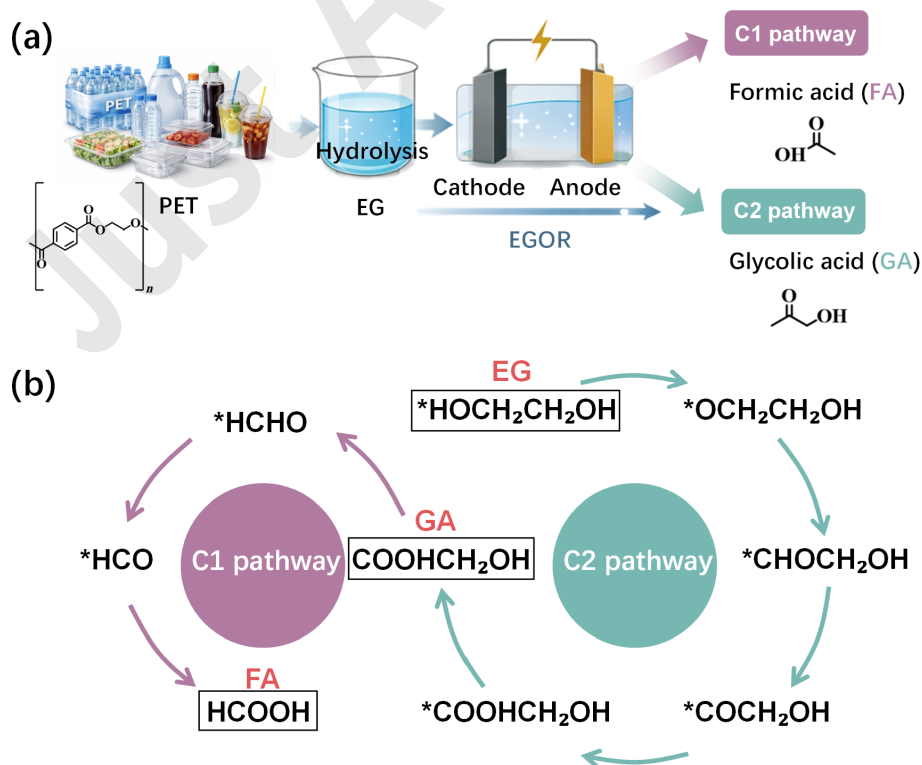
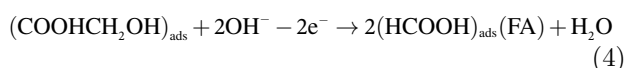
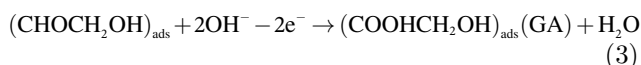
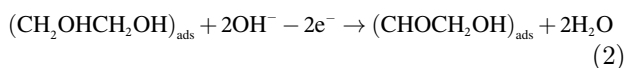
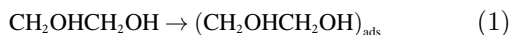


Figure 2 (a) Schematic illustration of PET hydrolysis to EG, followed by EGOR through two dominant pathways: the C1 pathway to FA and the C2 pathway to GA. (b) Schematic of EG electro-oxidation showing the competing C1 and C2 pathways leading to FA and GA.

oxidized to glycolaldehyde (CHOCH_2OH) and GA as early intermediates [29]. Further oxidation leads to C–C bond cleavage, generating C1 products (Fig. 2(b)). The C1 pathway follows a sequential surface reaction route of $^*\text{HOCH}_2\text{CH}_2\text{OH} \rightarrow ^*\text{CHOCH}_2\text{OH} \rightarrow ^*\text{COOHCH}_2\text{OH} \rightarrow ^*\text{HCOOH}$ [32]. The reaction equations leading to C1 products are presented in Eqs. (1)–(4) (Fig. 2(b)) [33–35]



High-valence metal oxyhydroxides, such as NiOOH and CoOOH , are commonly reported to promote the C1 pathway [36]. These materials can generate strong oxidative species and facilitate dehydrogenation and C–C bond breaking. The participation of lattice oxygen or surface hydroxyl species has also been proposed to lower the activation barrier for C–C cleavage. As a result, catalysts with strong oxidizing ability often show higher FA production but also a higher tendency toward over-oxidation. Qi et al. [33] used density functional theory (DFT) calculations to elucidate the reaction energetics of EGOR to FA. The free energy profiles show that EG is first adsorbed on NiOOH and then undergoes stepwise dehydrogenation to form glycolaldehyde, GA, formaldehyde, and finally FA. The rate-determining step differs between catalysts: For $\text{Cr}_{0.15}\text{-Ni}(\text{OH})_2\text{-NiOOH}$, it is formaldehyde dehydrogenation (1.18 eV), whereas for $\text{Cr-Ni}_3\text{S}_2\text{-NiOOH}$, it shifts to EG adsorption with a much lower barrier (0.67 eV). This indicates that sulfuration markedly facilitates EG adsorption and lowers the overall reaction barrier, while the synergistic effect of Cr and S stabilizes the NiOOH active phase and enhances catalytic activity.

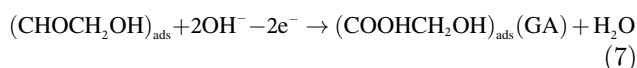
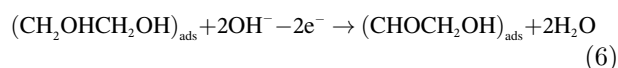
The applied potential plays a critical role in determining whether the C1 pathway is favored. At higher anodic potentials, the driving force for C–C bond cleavage increases, which promotes the formation of FA. However, excessively high potential also accelerates the OER, which competes with EG oxidation and reduces Faradaic efficiency toward organic products. Therefore, the C1 pathway is often associated with a narrow potential window in which FA formation is maximized while OER and complete mineralization are suppressed.

From a critical perspective, many reported C1-selective systems rely on relatively high overpotentials (η), which raises concerns about energy efficiency. Although FA is a valuable product, the energetic cost associated with deep oxidation may offset part of the benefit of replacing OER. Future catalyst design should aim to decouple C–C bond cleavage from excessive over-oxidation, enabling selective FA formation at lower potentials and with reduced energy input.

2.1.2 C2 pathway: EG to GA

The C2 pathway involves the selective EGOR without breaking the C–C bond and proceeds mainly through stepwise dehydrogenation to GA as the primary product (Fig. 2) [37, 38]. In the C2 pathway, EG is first oxidized to glycolaldehyde, followed

by further oxidation to GA [24, 39]. The reactions leading to C2 products are summarized in Eqs. (5)–(7) (Fig. 2(b)) [24, 34, 35, 39]



By avoiding C–C bond cleavage and requiring fewer oxidative and electron-transfer steps, this route is generally more thermodynamically and energetically favorable than the C1 pathway and is therefore typically preferred under milder oxidation conditions [40].

Catalysts that favor moderate oxidation strength are more likely to promote the C2 pathway. Precious metal catalysts with weaker oxidizing character have been reported to show higher selectivity toward GA. Zheng et al. [41] reveal that the Ir-doped catalyst maintains a high and stable FE (~90%) over 10 h for GA, while FE for FA remains suppressed, suggesting that Ir promotes the C2 pathway (GA) and inhibits C–C scission. Zhou et al. [42] provided a representative example of this principle using a mesoporous high-entropy alloy on nickel foam (m-HEA/NF). DFT-optimized geometries reveal that EGOR proceeds on m-HEA/NF through a sequence of surface-bound intermediates ($^*\text{HOCH}_2\text{CH}_2\text{OH} \rightarrow ^*\text{OCH}_2\text{CH}_2\text{OH} \rightarrow ^*\text{CHOCH}_2\text{OH} \rightarrow ^*\text{COCH}_2\text{OH} \rightarrow ^*\text{COOHCH}_2\text{OH}$), indicating a gradual dehydrogenation pathway that preserves the C–C bond toward GA formation. The corresponding free-energy profiles show that m-HEA/NF lowers the energetic requirements of the major uphill steps along the EG-to-GA pathway compared with m-Pd/NF. On m-HEA/NF, the rate-determining step is the dehydrogenation of $^*\text{COCH}_2\text{OH}$ to $^*\text{COOHCH}_2\text{OH}$, with a free-energy increase of 0.487 eV, which is lower than that of the same step on m-Pd/NF. In contrast, the rate-determining step on m-Pd/NF occurs during the conversion of $^*\text{HOCH}_2\text{CH}_2\text{OH}$ to $^*\text{OCH}_2\text{CH}_2\text{OH}$, requiring 0.686 eV, which is also higher than the corresponding value on m-HEA/NF. These results indicate that the multicomponent alloy reduces the major thermodynamic bottlenecks along the C2 pathway, thereby facilitating selective GA formation.

The applied potential is again a key parameter. At relatively low to moderate anodic potentials, the driving force is sufficient for dehydrogenation but not strong enough to induce extensive C–C bond cleavage. Under these conditions, GA selectivity is typically maximized. Increasing the potential often shifts the product distribution toward C1 products or even CO_2 , indicating a transition from selective oxidation to deep oxidation.

From a critical viewpoint, the C2 pathway is attractive from both energy and carbon-efficiency perspectives. By preserving the C–C bond, more carbon is retained in value-added products, and less energy is consumed per mole of product formed. However, GA separation and purification may introduce additional downstream costs. Therefore, although the C2 pathway is energetically favorable, its overall techno-economic advantage depends on both electrochemical selectivity and downstream processing considerations.

2.2 Why couple EGOR with HER?

Replacing the OER with EGOR fundamentally alters the

thermodynamic and kinetic landscape of electrolysis systems. Conventional water splitting requires an anodic equilibrium potential of 1.23 V vs. reversible hydrogen electrode (RHE) for OER, which is further increased by substantial kinetic overpotentials due to the four-electron transfer mechanism and sluggish O–O bond formation steps [43]. As a result, industrial alkaline electrolyzers typically operate above 1.6–1.8 V to achieve practical current densities for OER [30, 44].

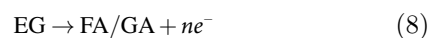
In contrast, EGOR proceeds at significantly lower anodic potentials because alcohol oxidation avoids the energetically demanding O–O bond formation step. For example, Liu et al. [30] reported that replacing the OER with EGOR significantly reduces the anodic operating potential in alkaline media. The Pt₁/RuO₂ catalyst requires 1.41 V vs. RHE to reach 10 mA·cm⁻² for OER, whereas the same current density can be achieved at potentials at least 800 mV lower during EGOR. In the two-electrode membrane-electrode assembly system, the EGOR/HER configuration reaches 100 mA·cm⁻² at only 1.14 V, while conventional OER/HER water splitting requires 2.35 V under identical conditions. Collectively, these studies highlight that EGOR substitution reduces thermodynamic requirements, lowers kinetic barriers, and enables co-production of chemicals and hydrogen, offering a compelling alternative to conventional OER-driven electrolysis systems.

2.3 Coupling mechanism of EGOR and HER

To improve the overall energy efficiency (EE_{cell}) of electrochemical systems, the anodic EGOR can be rationally coupled with various cathodic reduction reactions, including the HER, the 2e⁻ ORR to H₂O₂, CO₂RR, and NO₃⁻RR. These paired configurations enable simultaneous generation of value-added products at both electrodes while avoiding the sluggish OER. Among them, coupling EGOR with HER represents the most straightforward and practically relevant pathway because HER features fast kinetics [45], well-established catalyst systems, high product value, and simple gas–liquid separation, making it particularly suitable for evaluating the intrinsic energetic advantage of EGOR-based paired electrolysis.

As illustrated in Fig. 3, the anode selectively converts EG into value-added C1 and C2 products, such as FA and GA, in EGOR-coupled electrolysis, while the cathode produces H₂ through water reduction.

The anodic reaction can be generally expressed as [46]



where n is the number of electrons involved in the overall reaction.

In cathodic reaction, HER proceeds via the following reaction [47]



This configuration simultaneously realizes energy saving, hydrogen production, and PET-derived feedstock upgrading within a single electrochemical platform. In addition, the generation of gaseous hydrogen at the cathode and liquid organic products at the anode facilitates product separation, further enhancing the practical feasibility of the EGOR||HER coupling strategy.

2.4 Thermodynamic and energetic fundamentals

To translate EGOR||HER system into practical performance, a unified set of thermodynamic and energetic descriptors is required. The overall feasibility of EGOR||HER paired electrolysis is ultimately governed by how efficiently electrical energy is converted into chemical energy stored in hydrogen and value-added organic products. The key system-level parameters considered in EGOR||HER electrolysis include cell potential (E_{cell} (V)), η , FE, and EE_{cell} [48].

2.4.1 E_{cell}

Paired EGOR||HER electrolysis fundamentally redefines the energy landscape of water electrolysis by replacing the kinetically and thermodynamically demanding OER with the oxidation of EG. The overall E_{cell} is determined by the potential difference between the cathodic and anodic half-reactions [48, 49]

$$E_{\text{cell}} = E_{\text{anode}} - E_{\text{cathode}} \quad (10)$$

where E_{anode} is the equilibrium potential of the anodic reaction (V), and E_{cathode} is the equilibrium potential of the cathodic reaction (V), with both electrode potentials expressed relative to the same reference electrode.

In conventional water splitting, the anodic equilibrium potential for OER is 1.23 V vs. RHE under standard conditions. In contrast, EG oxidation to value-added C2 or C1 products proceeds at significantly lower thermodynamic potentials, depending on the product pathway. Consequently

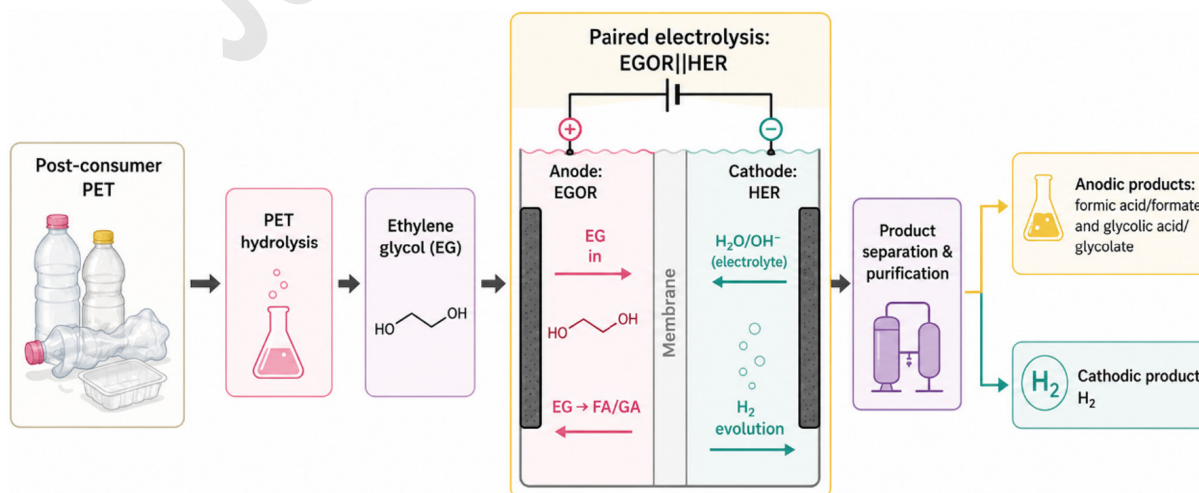


Figure 3 Schematic illustration of PET upcycling via EGOR||HER paired electrolysis and product separation.

$$E_{\text{cell}}^{\text{EGOR}} < E_{\text{cell}}^{\text{OER}} \quad (11)$$

where $E_{\text{cell}}^{\text{EGOR}}$ is the thermodynamic cell potential required for EGOR coupled with HER, and $E_{\text{cell}}^{\text{OER}}$ is the corresponding thermodynamic cell potential for conventional OER coupled with HER. This lower thermodynamic cell potential provides the fundamental basis for reducing the electrical energy input required for hydrogen production.

However, the practical operating voltage exceeds the thermodynamic value due to kinetic and resistive losses

$$E_{\text{cell,op}} = E_{\text{cell,thermo}} + \eta_{\text{anode}} + \eta_{\text{cathode}} + iR \quad (12)$$

where $E_{\text{cell,op}}$ is the practical operating cell voltage, $E_{\text{cell,thermo}}$ is the thermodynamic cell potential, η_{anode} is the anodic overpotential associated with EGOR, η_{cathode} is the cathodic overpotential associated with HER, i is the operating current, and R is the total Ohmic resistance of the cell. The iR term therefore represents the Ohmic voltage loss arising from ionic resistance in the electrolyte and membrane, as well as electronic and interfacial contact resistances. While EGOR typically exhibits a lower anodic overpotential than OER on optimized catalysts, operation at high current densities introduces additional voltage losses associated with charge-transfer limitations, mass transport resistance, and gas-bubble accumulation.

2.4.2 η

From a kinetic perspective, η originates from finite exchange current density and reaction barriers, commonly described by Butler–Volmer or Tafel relationships [14, 50, 51]

$$\eta = a + b \log j \quad (13)$$

where a is the Tafel intercept, b is the Tafel slope, and j is the current density. In paired EGOR systems, catalyst design must simultaneously optimize anodic alcohol dehydrogenation and cathodic HER kinetics, as the total overpotential is additive

$$\eta_{\text{total}} = \eta_{\text{anode}} + \eta_{\text{cathode}} \quad (14)$$

where η_{total} is the total kinetic overpotential of the paired electrolyzer, η_{anode} is the anodic overpotential associated with EGOR, and η_{cathode} is the cathodic overpotential associated with HER.

2.4.3 FE

FE quantifies the fraction of total charge that is used to form a specific desired product. It is defined as [46, 52]

$$\text{FE} = \frac{nF N_{\text{product}}}{Q_{\text{total}}} \times 100\% \quad (15)$$

where F is the Faraday constant ($96,485 \text{ C}\cdot\text{mol}^{-1}$), N_{product} is the number of moles of product formed, and Q_{total} is the total charge passed.

High FE toward target anodic products (FA or GA) indicates efficient utilization of electrons for EG oxidation. At the same time, FE losses can arise from competing reactions, such as OER at the anode or side reactions at the cathode. From a system perspective, both anodic and cathodic FE values should be considered, because low FE at either electrode directly reduces the overall effectiveness of paired electrolysis.

2.4.4 EE_{cell}

The overall EE_{cell} reflects how effectively electrical energy is

converted into chemical energy stored in the products. A commonly used definition is [14, 53]

$$EE_{\text{cell}} = \frac{\Delta G_{\text{products}}}{E_{\text{cell,op}} nF} \times \text{FE} \quad (16)$$

where $\Delta G_{\text{products}}$ is the Gibbs free energy change associated with formation of the desired products, and $E_{\text{cell,op}}$ is the operating cell voltage.

Alternatively, EE_{cell} can be expressed in terms of electrical energy input per amount of product

$$\text{Energy consumption} = \frac{E_{\text{cell,op}} It}{m_{\text{product}}} \quad (17)$$

where I is the current, t is the operating time, and m_{product} is the mass of product formed.

In paired EGOR systems, lowering $E_{\text{cell,op}}$ through substitution of OER with EGOR, together with high FE toward value-added anodic products, can substantially improve EE_{cell} compared with conventional water electrolysis. Critically, optimizing a single parameter, such as minimizing overpotential or maximizing FE, does not necessarily maximize EE_{cell} . Instead, balanced optimization of cell voltage, selectivity, and operating current density is required to achieve truly energy-efficient paired EGOR systems.

3 Catalyst design for EGOR||HER coupled electrolysis

To enable efficient and practical EGOR, catalyst design must be considered from a system-level perspective rather than for a single electrode. In a paired EGOR||HER electrolyzer, the anode and cathode always operate in the same electrolyte and under identical operating conditions, which requires the two electrodes to be electrochemically compatible [19, 20]. Notably, recent studies reveal a substantial overlap in the material requirements for EGOR and HER catalysts [54, 55]. Catalyst families that are active for EGOR, such as oxyhydroxides, chalcogenides, phosphides, nitrides, and carbon [56], are in most cases also intrinsically active for alkaline HER [9, 57]. This intrinsic convergence makes it possible for a single material platform to drive both half-reactions.

As a direct consequence, many reported EGOR||HER systems adopt symmetric or quasi-symmetric configurations, in which identical or closely related catalysts are used at both electrodes. Such a design minimizes material incompatibility, simplifies device fabrication, ensures matched reaction environments, and improves long-term operational stability [21]. From a practical engineering perspective, symmetric systems therefore represent the most straightforward pathway toward integrated electrolysis devices.

In addition, the anode and cathode can be equipped with individually optimized catalysts for EGOR and HER in asymmetric configurations [57]. This asymmetric pairing enables independent regulation of anodic oxidation strength, hydrogen-binding energetics, and surface reconstruction behavior, thereby improving intrinsic cell performance. However, using two completely different catalysts in a single industrial electrolyzer also increases manufacturing complexity, electrode fabrication cost, and quality-control requirements. In addition, the two electrodes may show different degradation rates, reconstruction behaviors, impurity tolerance, and maintenance needs, leading to long-term stability mismatch at the device level. Therefore, asymmetric systems should be evaluated not only by initial activity, but also by

scalability, catalyst cost, electrode lifetime balance, and compatibility under continuous operation.

The fundamental origin of this catalyst convergence lies in their shared mechanistic requirements. Both EGOR and HER rely on balanced surface adsorption energetics: Catalysts that bind intermediates neither too weakly nor too strongly tend to be effective for both reactions. For EGOR, moderate binding of oxygenated intermediates (e.g., CHO and COCH₂OH) is required to avoid surface poisoning, whereas for HER, an optimal hydrogen binding energy (near thermoneutral hydrogen adsorption free energy (ΔG_{H^+})) is essential for fast H–H coupling or electrochemical desorption. In addition, both reactions benefit from high electrical conductivity and fast charge-transfer kinetics, which are common features of metals, alloys, and many transition-metal compounds. Moreover, similar electronic descriptors, such as the d-band center and local charge redistribution, govern the activity of both processes: Tuning the d-band downward weakens overly strong adsorbate binding for EGOR while simultaneously bringing hydrogen adsorption closer to the optimum for HER.

Therefore, catalyst selection for coupled EGOR||HER should be understood as a balance between system integration and intrinsic catalytic performance. Symmetric systems provide the most device-compatible and scalable configuration, whereas asymmetric systems offer a pathway toward the highest attainable activity and energy efficiency through independent optimization of each half-reaction.

This section focuses on bifunctional or mutually compatible catalysts that can simultaneously support both EGOR and HER (Fig. 4). We highlight representative material classes, key design principles, and structure–property relationships that govern activity, selectivity, and durability in coupled EGOR||HER systems, providing guidelines for next-generation catalyst development for integrated plastic valorization and green hydrogen production.

3.1 Precious metal catalysts

Single precious-metal catalysts, such as Pt and Pd, play a central mechanistic role in EGOR by providing a balanced adsorption strength for EG and intermediates, enabling fast dehydrogenation without persistent surface poisoning [58]. Under anodic

polarization, their surfaces undergo controlled oxidation, generating metal-oxygen species that promote C–H activation and tune the competition between C–C cleavage and C–C retention pathways [59–61]. Because these surface states evolve predictably with potential, they allow direct correlation among applied potential, intermediate evolution, and FE, especially for C₂ products. Importantly, the same Pt- and Pd-based surfaces also exhibit excellent activity for the HER in alkaline media, with favorable hydrogen adsorption/desorption energetics and low overpotentials. This intrinsic bifunctionality makes single precious metals a useful benchmark for coupled EGOR||HER systems, where catalyst performance must be evaluated not only by anodic selectivity but also by cathodic hydrogen-production efficiency [62].

Doping is a significant strategy to enhance single precious-metal catalysts for coupled EGOR||HER systems [24, 38, 63]. Although single precious-metal catalysts have demonstrated high activity and selectivity in EGOR, their further advancement is constrained by both intrinsic electronic rigidity and economic considerations [62, 64]. In single-metal systems, the adsorption energetics of EG, hydroxyl species, and oxygenated intermediates are governed by the fixed electronic structure of the noble metal, limiting the tunability of reaction pathways, potential-dependent selectivity, and overall paired-electrolysis performance [41, 65, 66]. Meanwhile, the reliance on high precious-metal loadings substantially increases material cost and hinders large-scale deployment [40, 67].

Introducing transition metals via doping or alloying offers a rational route to simultaneously address these performance and cost limitations in bifunctional EGOR||HER catalysis [65]. Mechanistically, incorporating secondary metals enables precise modulation of the local electronic structure, such as charge redistribution and d-band center tuning, which optimizes the adsorption of EGOR intermediates [38] while also adjusting hydrogen binding strength for efficient HER. From a practical standpoint, partial substitution of precious metals with earth-abundant transition metals directly reduces noble-metal usage without sacrificing catalytic efficiency. More importantly, synergistic interactions in alloy catalysts often create new or modified active sites that outperform single-metal counterparts, enabling higher FE for EGOR, lower operating potentials, and improved HER kinetics at reduced precious-metal content.

Zheng et al. [41] proposed that Ir doping in multicomponent high-entropy alloy nanoparticles (Ir–PdPtAuNiCu HEANs) plays a decisive role in enhancing coupled EGOR||HER performance. As shown in Fig. 5(a), Ir incorporation markedly lowers the EGOR onset potential and significantly increases mass activity in 1 M NaOH + 0.5 M EG, delivering the highest current at 0.724 V vs. RHE. Figure 5(b) further reveals superior durability: After 1000 potential cycles, the Ir-doped HEANs retain nearly identical polarization behavior, whereas the undoped HEANs show noticeable decay. On the cathodic side, Fig. 5(c) shows that Ir-HEANs also exhibit faster HER kinetics with lower overpotentials and steeper polarization curves, confirming that the same doping strategy benefits both anodic EGOR and cathodic HER. The flow electrolyzer (Fig. 5(d)) illustrates a membrane-free configuration of (–)PdPtAuNiCu||Ir–PdPtAuNiCu(+), enabling stable continuous operation with efficient anodic EG-to-GA conversion coupled to high-rate HER.

Zhang et al. [29] demonstrated that doping precious metals with transition metals is an effective strategy to develop bifunctional catalysts for coupled EGOR||HER. As shown

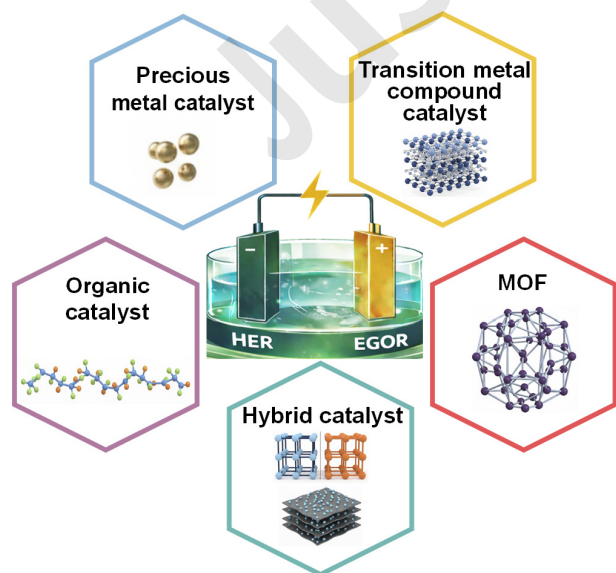


Figure 4 Schematic illustration of catalyst design for coupled EGOR||HER systems.

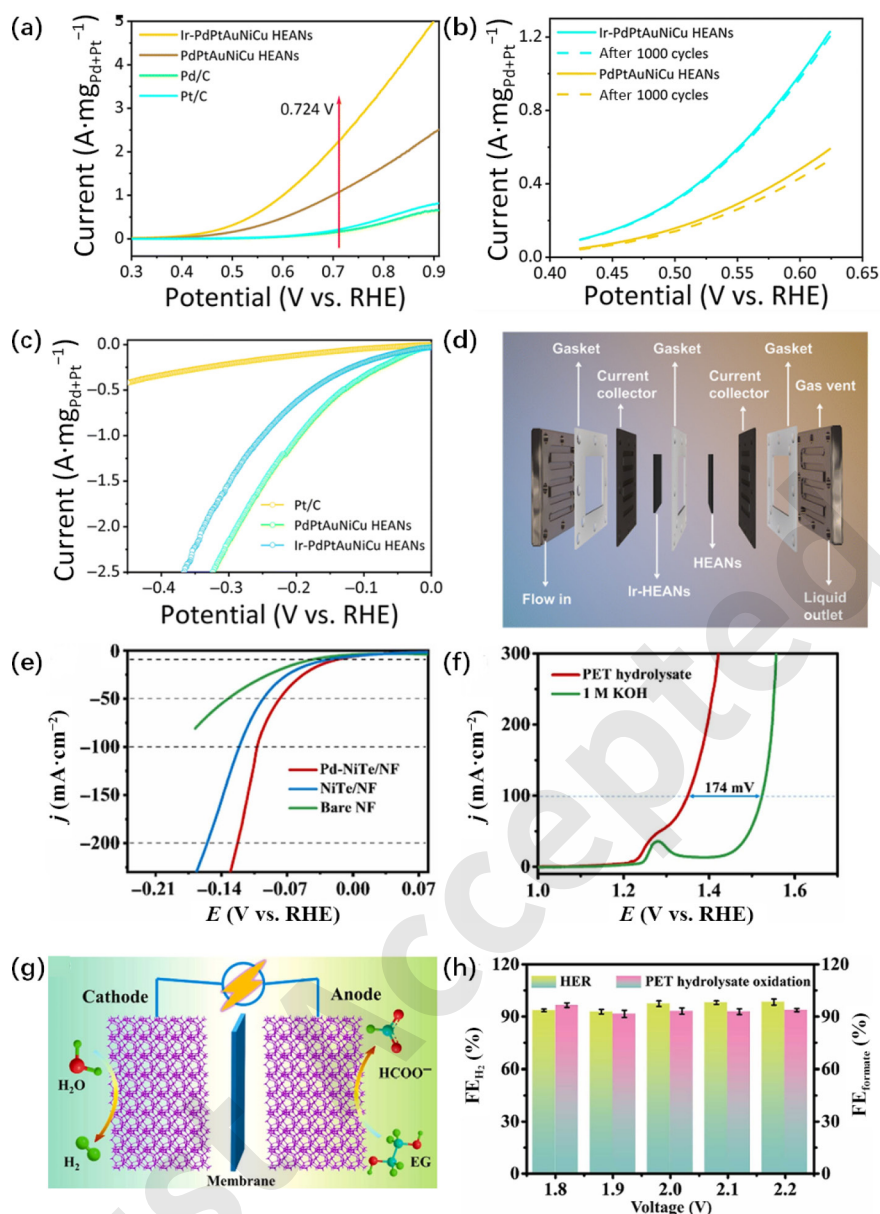


Figure 5 (a) Linear sweep voltammetry (LSV) curves of Ir-PdPtAuNiCu HEA nanoparticles (Ir-HEANs), PdPtAuNiCu HEANs, and commercial Pt/C and Pd/C in 1 M NaOH + 0.5 M EG at 5 mV·s⁻¹. (b) EGOR stability comparison of Ir-HEANs and PdPtAuNiCu HEANs before and after 1000 potential cycles. (c) HER LSV curves of Ir-HEANs, PdPtAuNiCu HEANs, and Pt/C in 1 M NaOH at 5 mV·s⁻¹. (d) Schematic illustration of a membrane-free flow electrolyzer configured as (-)PdPtAuNiCu||Ir-PdPtAuNiCu(+) for paired EGOR||HER. Reproduced with permission from Ref. [41], © The Royal Society of Chemistry 2025. (e) HER polarization curves of Pd-NiTe/NF, NiTe/NF, and bare NF in 1 M KOH. (f) LSV curves of Pd-NiTe/NF of EGOR in 1 M KOH. (g) Schematic illustration of EGOR||HER using Pd-NiTe/NF as cathode and anode. (h) Faradaic efficiencies for H₂ generation at the cathode and FA production at the anode in EGOR||HER cell. Reproduced with permission from Ref. [29], © Elsevier B.V. 2023.

in Fig. 5(e), Pd-NiTe/NF exhibits a lower HER overpotential, indicating improved HER kinetics. Meanwhile, Fig. 5(f) reveals a reduced oxidation onset potential in PET hydrolysate, suggesting that Ni-Te doping facilitates EG activation and suppresses competing OER. The paired electrolysis configuration in Fig. 5(g) illustrates that Pd-NiTe/NF can simultaneously drive anodic EGOR to formate and cathodic HER in a membrane-separated cell. Furthermore, Fig. 5(h) shows that both H₂ and formate maintain Faradaic efficiencies above 90% across 1.8–2.2 V, highlighting the high selectivity and stability of this doped precious-metal catalyst system.

3.2 Transition metal compound catalysts

Although precious-metal catalysts generally exhibit high intrinsic

activity and well-defined reaction behavior for EGOR, their high cost and limited abundance strongly restrict large-scale application. Transition-metal catalysts offer clear economic advantages; however, monometallic systems often suffer from rigid electronic structures, insufficient stability, and limited control over reaction selectivity, while also failing to deliver balanced performance for both EGOR and HER [68]. To address these limitations, increasing attention has been directed toward transition-metal compounds with more diverse compositions and structures that can serve as bifunctional catalysts. Materials, such as layered double hydroxides (LDHs), oxides, sulfides, selenides, phosphides, nitrides, carbides, and carbonitrides (MXenes) [69], provide highly tunable electronic states and surface chemistries. Through rational regulation of composition, phase, and local

structure, these materials enable simultaneous optimization of anodic EGOR activity/selectivity and cathodic HER kinetics, while improving structural durability. Consequently, transition-metal compounds represent promising, cost-effective alternatives to precious-metal-based catalysts for coupled EGOR||HER systems.

3.2.1 Oxyhydroxides

Transition-metal oxyhydroxides ($-\text{OOH}$) are effective catalysts for EGOR due to their rich redox chemistry and hydroxyl-rich surfaces under anodic conditions. During operation, high-valence metal centers are dynamically generated, providing active sites that promote EG dehydrogenation through metal-OH-assisted pathways [70]. Meanwhile, the reversible redox transformation of oxyhydroxides enables continuous regeneration of active sites and stable surface chemistry, which supports sustained catalytic activity during EGOR. Importantly, these same oxyhydroxide surfaces can also facilitate the HER under cathodic polarization by providing favorable sites for water dissociation and hydrogen adsorption/desorption. Thus, transition-metal oxyhydroxides can serve as bifunctional catalysts that simultaneously support efficient EGOR at the anode and fast HER at the cathode in coupled EGOR||HER systems.

Wu et al. [70] developed a reconstructed Ni^{2+} -doped CoOOH nanosheet array as a representative oxyhydroxide catalyst for EGOR derived from PET hydrolysate. The catalyst was obtained via electrochemical reconstruction from a Ni^{2+} - $\text{Co}(\text{OH})_2$ precursor, forming ultrathin CoOOH nanosheets uniformly anchored on nickel foam, which maximized active site exposure and ensured efficient electron transport. Ni^{2+} incorporation modulates the local electronic structure of CoOOH , enhances EG adsorption, and accelerates the generation of reactive $^*\text{OH}$ species, thereby facilitating C-C bond cleavage during EGOR. As a result, the optimized Ni^{2+} - CoOOH catalyst achieves industrial-level performance, delivering $500 \text{ mA}\cdot\text{cm}^{-2}$ at 1.38 V vs. RHE with $> 90\%$ FE for formate over a wide current density range, highlighting the intrinsic advantages of oxyhydroxide materials with dynamically regenerating active surfaces for high-rate EGOR.

3.2.2 LDHs

Following transition-metal oxyhydroxides, LDHs, such as Ni-LDH, Co-LDH, and Fe-LDH, have emerged as versatile oxide-based catalysts for both EGOR and HER owing to their composition-tunable metal layers and hydroxyl-rich interlayers [26, 71]. Under anodic potentials, LDHs undergo surface reconstruction to generate high-valence metal (oxy)hydroxide species that mediate metal-OH-assisted EG dehydrogenation and C-C bond cleavage [6, 70]. Meanwhile, under cathodic conditions, their hydroxyl-rich layered structure facilitates water dissociation, while optimized metal compositions regulate hydrogen adsorption energy, enabling efficient alkaline HER [31, 72]. The layered architecture promotes hydroxyl adsorption and ion transport, and multimetal synergy fine-tunes intermediate binding and reaction selectivity for EGOR while simultaneously accelerating Volmer kinetics for HER [48, 71, 73, 74]. Lu et al. [71] reported a cobalt-vanadium LDH (CoV-LDH) oxide catalyst as an efficient anode for EGOR derived from PET hydrolysate. Benefiting from the multimetal LDH framework and tunable electronic structure, the incorporation of high-valence V species promotes the *in situ* formation of Co^{3+} active sites, enhances OH^- adsorption, and accelerates lattice-oxygen-assisted dehydrogenation of EG. As a result, CoV-LDH delivers an industrial-level current density of $350 \text{ mA}\cdot\text{cm}^{-2}$ at 1.47 V (vs.

RHE) with a high FA selectivity of 86.61%. Mechanistic studies combining electrochemical analysis and *in situ* Fourier-transform infrared (FTIR) spectroscopy reveal that EG oxidation proceeds via a $\text{Co}^{2+}/\text{Co}^{3+}$ redox-mediated pathway, where EG is selectively converted to FA through C-C bond cleavage on Co^{3+} -OH active centers, highlighting the intrinsic advantages of LDH-type oxide catalysts for selective and energy-efficient EGOR. Lu et al. [49] constructed an LDH-derived $\text{PtPd-Ni}(\text{OH})_2$ catalyst by anchoring alloyed PtPd nanoparticles on $\text{Ni}(\text{OH})_2$ nanosheets. The $\text{Ni}(\text{OH})_2$ layered structure supplies abundant $^*\text{OH}$ species and $\text{Ni}^{2+}/\text{Ni}^{3+}$ redox sites to mediate EG dehydrogenation, while PtPd alloying optimizes intermediate adsorption and stabilizes metallic sites. The catalyst requires only 0.69 V vs. RHE to reach $200 \text{ mA}\cdot\text{cm}^{-2}$ and achieves 90.3% FE for GA with stability over 700 h. Integrated into a HER||EGOR electrolyzer, the system significantly lowers cell voltage compared to water splitting, enabling energy-efficient H_2 production alongside PET-derived EG upgrading. This work highlights LDH-based catalysts as robust bifunctional platforms for high-rate EGOR and paired HER.

3.2.3 Oxides

Transition-metal oxides, such as NiO [75], Co_3O_4 [22, 27, 76], and NiCo_2O_4 [53, 77, 78], constitute another important class of catalysts for both EGOR and HER owing to their robust crystal frameworks and versatile redox chemistry in alkaline media. For EGOR, the coexistence of variable metal valence states and abundant surface oxygen species facilitates hydroxyl activation and enables EG dehydrogenation via lattice-oxygen or metal-oxygen-hydroxyl pathways [79, 80]. Meanwhile, under cathodic conditions, certain oxide surfaces promote water dissociation and stabilize hydrogen adsorption intermediates, contributing to improved alkaline HER kinetics. Compared with layered hydroxides, their more rigid oxide lattices better preserve active sites under prolonged anodic operation, mitigate catalyst poisoning, and maintain structural integrity during high-current electrolysis [36, 78, 81, 82]. Consequently, transition-metal oxides provide a complementary platform to LDHs, balancing catalytic efficiency for EG oxidation with durability and operational stability in integrated EGOR||HER paired systems. Xie et al. [83] reported a series of Cu/Sn -based medium-entropy alloy oxides (MEAOs) as efficient oxide catalysts for EGOR coupled with hydrogen evolution. By constructing multimetal oxide lattices, these MEAOs exhibit enhanced redox flexibility and strong metal-oxygen interactions, which are favorable for both EGOR at the anode and HER at the cathode. Among them, $\text{Cu}_{0.5}\text{Co}_{0.5}\text{SnO}_{3.17}$ showed the most favorable catalytic performance, delivering a low HER overpotential of 181 mV at $10 \text{ mA}\cdot\text{cm}^{-2}$ and enabling overall electrolysis at a reduced cell voltage of 1.26 V , outperforming the commercial $\text{RuO}_2 + \text{Pt/C}$ system. In addition, this oxide catalyst achieved a high FE of 97.7% for FA formation during EGOR at 1.6 V . Density functional theory analysis revealed that the superior activity originates from a d-band center closer to the Fermi level and strong Co 3d-Cu/Sn orbital synergy, highlighting the critical role of electronic structure regulation in multimetal oxide catalysts.

3.2.4 Sulfides

While transition-metal oxides provide structural robustness and well-defined redox chemistry for EGOR, their relatively poor electrical conductivity often limits performance at high current densities and can also restrain alkaline HER kinetics due to sluggish electron transfer. By contrast, transition-metal sulfides,

such as Ni_3S_2 [84] and Co_9S_8 [18], possess a more metallic electronic structure and higher intrinsic conductivity, enabling faster charge transport under both anodic and cathodic polarization. Under alkaline conditions, sulfides readily undergo *in situ* surface reconstruction to generate catalytically active oxyhydroxide species that drive EGOR [33, 85], while their conductive cores facilitate rapid electron delivery for HER. In addition, the reconstructed interfaces often enhance water dissociation and optimize hydrogen adsorption, promoting efficient alkaline HER. Consequently, transition-metal sulfides provide a dynamic and bifunctional platform that supports high-rate EGOR while simultaneously improving HER performance in integrated paired electrolysis systems.

In this context, Pataniya et al. [85] reported a cerium-doped nickel-cobalt sulfide (Ce-NiCoS) catalyst directly grown on nickel foam via one-step chemical bath deposition for paired PET electro-upcycling and hydrogen evolution. The conductive NiCo_2S_4 backbone enabled fast electron transport, while Ce incorporation promoted surface reconstruction to *in situ* formed CoOOH/NiOOH , which served as the true active phase for indirect EG oxidation. As a result, the Ce-NiCoS anode reached $100 \text{ mA}\cdot\text{cm}^{-2}$ at 1.30 V vs. RHE for EGOR (230 mV lower than OER) with 96.5% FE toward formate and enabled bifunctional water/PET hydrolysate electrolysis with excellent stability at industrial current densities. This work illustrates how rationally engineered sulfides can bridge the gap between oxide chemistry and highly conductive chalcogenides to achieve efficient and durable PET electro-upgrading.

3.2.5 Selenides

Building on the advances of transition-metal sulfides, which already offer improved conductivity over oxides and hydroxides, transition-metal selenides, such as NiSe_2 and CoSe_2 , have emerged as an even more conductive alternative for both EGOR and HER due to their more metallic character and higher lattice polarizability [86, 87]. For EGOR, the larger ionic radius and lower electronegativity of selenium weaken metal-chalcogen bonding, facilitate OH^- adsorption [88], and accelerate interfacial charge transfer, thereby promoting alcohol dehydrogenation and C–C bond cleavage in alkaline media [87].

Meanwhile, these electronic features also benefit HER: The weakened metal–Se interaction modulates the d-band structure, optimizes hydrogen adsorption energy, and enhances water dissociation kinetics in alkaline conditions. The improved conductivity ensures rapid electron delivery under cathodic polarization, while surface reconstruction can generate active oxyhydroxide or metal-rich sites that further accelerate hydrogen evolution. Consequently, transition-metal selenides provide a highly conductive and electronically tunable platform that simultaneously enhances EGOR and HER in paired electrolysis systems.

Sharma et al. [86] developed a binder-free hierarchically structured $\text{Ni-Fe}_3\text{Se}_4$ catalyst directly grown on nickel foam for coupled PET electro-upcycling and hydrogen production. The highly conductive selenide backbone reduced interfacial resistance and promoted the *in situ* formation of redox-active NiFeOOH , which acts as the true catalytic phase during EGOR. Consequently, the electrode reached $\sim 1.37 \text{ V}$ at $100 \text{ mA}\cdot\text{cm}^{-2}$ with an 89% FE toward formate at $50 \text{ mA}\cdot\text{cm}^{-2}$, outperforming many analogous sulfide systems under comparable conditions. Moreover, the same $\text{Ni-Fe}_3\text{Se}_4$ electrode sustained bifunctional water electrolysis for > 42 h at $300 \text{ mA}\cdot\text{cm}^{-2}$, illustrating how selenides can retain the

structural advantages of sulfides while delivering superior conductivity and reaction kinetics for scalable plastic electro-upgrading.

3.2.6 Phosphides

Metal phosphides, such as Ni_2P , CoP , FeP , and NiCoP , represent a further evolution of chalcogenide-based catalysts for EGOR, benefiting from even higher electrical conductivity and highly tunable metal-phosphorus bonding [9, 89]. Under anodic conditions, phosphide surfaces readily reconstruct into catalytically active metal (oxy)hydroxide species that mediate EG dehydrogenation through metal–OH-assisted pathways [25, 90]. Owing to their more metallic character, phosphide-derived catalysts generally enable faster charge transfer and lower kinetic barriers than conventional oxides or hydroxides, which not only enhances EGOR kinetics but also favors efficient hydrogen adsorption/desorption during HER [26, 91]. Consequently, metal phosphides typically deliver enhanced EGOR activity, stronger resistance to surface poisoning, and superior performance at high current densities, while simultaneously exhibiting favorable HER kinetics, making them attractive bifunctional catalysts for coupled EGOR||HER systems [92, 93].

Zhang et al. [9] fabricated a $\text{Co}_2\text{P-Ni}_2\text{P}$ heterostructure on carbon cloth ($\text{Co}_2\text{P-Ni}_2\text{P/CC}$) via plasma phosphating of electrodeposited CoNi-LDH nanosheets, preserving a porous vertical architecture with improved conductivity (Fig. 6(a)). In 1.0 M KOH containing 0.5 M EG, $\text{Co}_2\text{P-Ni}_2\text{P/CC}$ exhibited the lowest onset potential and the highest current density for EGOR among all control catalysts (CC, $\text{Co}_2\text{P/CC}$, $\text{Ni}_2\text{P/CC}$, CoNi-LDH/CC , $\text{CoNiO}_x\text{/CC}$, and $\text{RuO}_2\text{/CC}$) (Fig. 6(b)). The smaller Tafel slope ($\sim 126.6 \text{ mV}\cdot\text{dec}^{-1}$) indicates faster charge-transfer kinetics, attributable to the $\text{Co}_2\text{P-Ni}_2\text{P}$ heterointerface that facilitates OH^- activation and interfacial electron transport (Fig. 6(c)). The same catalyst also shows superior HER performance (Fig. 6(d)), enabling a symmetric $\text{Co}_2\text{P-Ni}_2\text{P/CC}$ || $\text{Co}_2\text{P-Ni}_2\text{P/CC}$ cell to operate at $\sim 306 \text{ mV}$ lower voltage than 20% Pt/C || RuO_2 at $100 \text{ mA}\cdot\text{cm}^{-2}$ (Fig. 6(e)). Overall, this work demonstrates that bimetallic phosphides are cost-effective, bifunctional catalysts for efficient coupled EGOR||HER.

3.2.7 Nitrides

Transition metal nitrides have recently emerged as promising non-noble electrocatalysts because of their high electrical conductivity, rapid charge-transfer kinetics, and more favorable surface electronic structures than conventional oxides or hydroxides [94–96]. Nitrogen incorporation not only increases metallic character but also redistributes electron density around metal centers, tailoring their coordination environment and surface reactivity [96]. This electronic modulation optimizes the adsorption and activation of key intermediates for both the HER and organic molecule oxidation, such as EGOR. N-induced tuning typically brings the ΔG_{H^+} closer to the thermoneutral value while simultaneously promoting the formation and reactivity of surface hydroxyl species required for alcohol oxidation [68, 95, 97]. Furthermore, multimetal doping and heterostructure engineering in nitrides can create more exposed active sites, strengthen structural robustness, and refine reaction selectivity. As a result, transition-metal nitrides are increasingly recognized as efficient bifunctional catalysts capable of simultaneously driving EGOR and HER in coupled electrolysis systems.

Liu et al. [68] reported that transition-metal nitride $\text{Co-Ni}_3\text{N/CC}$ is an effective bifunctional electrode for simultaneous

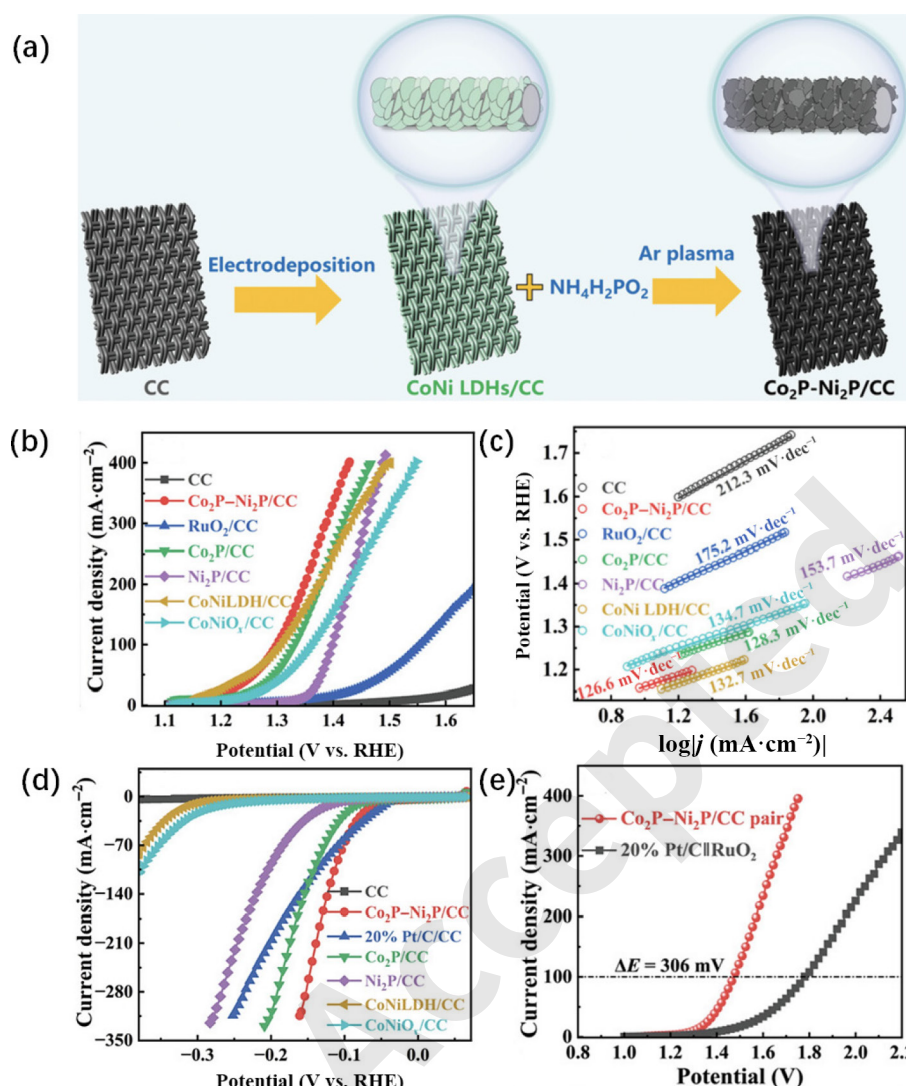


Figure 6 (a) Schematic illustration of the plasma phosphating synthesis of Co₂P-Ni₂P/CC. (b) LSV curves and (c) Tafel plots of CC, Co₂P-Ni₂P/CC, RuO₂/CC, Co₂P/CC, Ni₂P/CC, CoNi LDH/CC, and CoNiO_x/CC in 1.0 M KOH containing 0.5 M EG. (d) HER curves of CC, Co₂P-Ni₂P/CC, 20% Pt/C/CC, Co₂P/CC, Ni₂P/CC, CoNi LDH/CC, and CoNiO_x/CC in 1 M KOH. (e) LSV comparison of the Co₂P-Ni₂P/CC couple and 20% Pt/C||RuO₂ in 1 M KOH containing 0.5 M EG. Reproduced with permission from Ref. [9], © Wiley-VCH GmbH 2024.

PET upcycling and hydrogen production. As shown in Fig. 7(a), hydrothermally grown NiCo-OH nanosheets are converted into porous Co-Ni₃N via high-temperature nitridation; nitrogen incorporation imparts stronger metallic character, higher electrical conductivity, and more favorable surface charge distribution while preserving interconnected channels for mass transport and bubble release. Figure 7(b) reveals that, compared with Ni₃N/CC, Co-doped nitride exhibits a lower EGOR onset potential and higher current density in 1 M KOH + 0.5 M EG, while suppressing competing OER, indicating that the nitride surface promotes more selective EG oxidation. Figure 7(c) shows that Co-Ni₃N/CC delivers superior HER kinetics with a more positive onset potential and higher cathodic current than Ni₃N/CC, NiCo-OH precursor, and Pt/C, suggesting that the nitride surface provides near-optimal H adsorption/desorption. Based on this, a symmetric cell is constructed in Fig. 7(d) using Co-Ni₃N/CC as both anode and cathode for paired EGOR||HER. The full-cell curves in Fig. 7(e) further demonstrate that operating in PET hydrolysate significantly lowers the cell voltage, showing that a nitride-centered symmetric system enables both plastic valorization and improved energy efficiency. Overall, this work highlights

nitridation as a key strategy to enhance the bifunctional performance of transition-metal catalysts for energy-efficient paired electrolysis.

3.3 MOFs catalysts

MOFs have attracted growing interest in both EGOR and HER owing to their high surface area, tunable coordination environments, and ordered porosity, which facilitate reactant adsorption, intermediate stabilization, and mass transport [28, 31, 98–102]. In EGOR, the well-defined metal sites promote alcohol adsorption and redox cycling (e.g., Ni²⁺/Ni³⁺), while in HER, tailored coordination structures and heteroatom modulation can optimize hydrogen adsorption and accelerate water dissociation in alkaline media [28, 31, 98, 100]. Furthermore, hierarchical or template-confined MOF architectures are increasingly explored to mitigate nanosheet stacking, enhance electrical conductivity, and expose more accessible active sites, thereby improving practical bifunctional electrocatalytic performance in paired EGOR||HER systems [102]. In this context, Li et al. [102] developed a biocarbon-confined orthogonal nanosheet FeCo(MOF)@C catalyst via a yeast-templated strategy, in which bimetallic MOF nanosheets

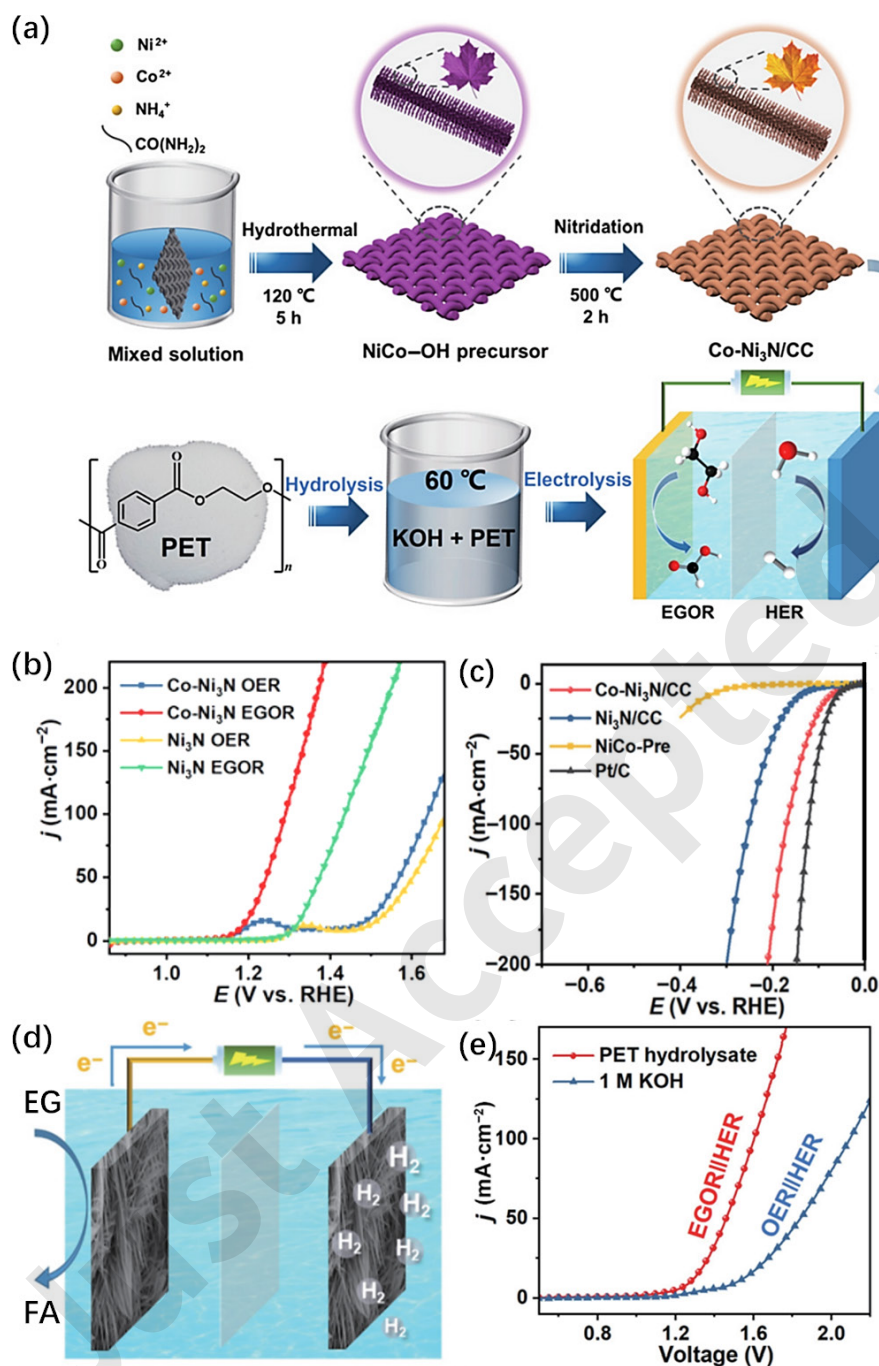


Figure 7 (a) Schematic illustration of the synthesis of Co-Ni₃N/CC and its application for simultaneous EGOR and HER. (b) LSV curves of Co-Ni₃N/CC and Ni₃N/CC in 1.0 M KOH with and without 0.1 M EG. (c) HER polarization curves of Co-Ni₃N/CC, Ni₃N/CC, NiCo-OH precursor, and Pt/C in 1 M KOH. (d) Schematic of the symmetric paired EGOR||HER electrolyzer employing Co-Ni₃N/CC as both anode and cathode. (e) Cell polarization curves of the paired Co-Ni₃N/CC system in PET hydrolysate versus pure 1 M KOH, demonstrating reduced cell voltage for EGOR||HER. Reproduced with permission from Ref. [68], © Tsinghua University Press 2022.

were orderly assembled on conductive carbon spheres rather than randomly stacked; this architecture enhanced electrochemically accessible surface area, lowered charge-transfer resistance, and strengthened EG adsorption at Fe/Co dual sites, enabling only 1.42 V to reach 100 mA·cm⁻² for EGOR, an HER overpotential of 307 mV at 100 mA·cm⁻², an overall EGOR||HER cell voltage of 1.83 V, and stable operation for > 160 h at 500 mA·cm⁻², thereby demonstrating that bio-templated MOF designs can reconcile high activity with industrial-level durability. Complementing this strategy, He et al. [103] further showed that coupling MOFs with noble metals can create highly active metal-MOF heterointerfaces:

They embedded Pt nanoclusters with exposed (111) facets into MIL-100(Fe), where abundant surface oxygen sites formed strong Pt-O interactions that promoted interfacial charge transfer, increased accessible Pt sites, and accelerated proton-electron coupling. Benefiting from this metal-support interaction, Pt/MIL-100(Fe) delivered exceptional bifunctional performance, including very low HER overpotentials (46/29 mV in acid/alkali) and outstanding EGOR activity with a mass activity of 11,826 mA·mg⁻¹, far surpassing commercial Pt/C. Moreover, the MOF scaffold effectively mitigated Pt poisoning and dissolution, affording superior long-term stability and anti-poisoning

capability during EGOR. Together, these studies illustrate that MOF-based electrocatalysts, whether carbon-confined or noble-metal-hybridized, offer a versatile platform to integrate high surface area, tailored electronic structure, and interfacial engineering for efficient and durable alcohol electro-oxidation.

3.4 Organic catalysts

Distinct from inorganic metal-based catalysts that rely on surface-bound metal–OH intermediates, organic electrocatalysts for EGOR mainly operate through molecular or outer-sphere electron-transfer pathways [104]. This category encompasses both soluble organic mediators and organic/coordination polymer-based catalysts, which share tunable molecular structures, well-defined redox chemistry, and high controllability of product distribution via electrolyte conditions.

In this framework, Behera et al. [19] demonstrated a cobalt-based one-dimensional coordination polymer that behaves as a molecular-like organic framework catalyst for PET electro-upcycling: It operated at a low onset potential of 1.27 V vs. RHE, enabled quantitative recovery of terephthalic acid and high selectivity to potassium diformate, and revealed that formate over-oxidation proceeds via a formate (rather than oxalate) pathway, highlighting the importance of potential control in such molecularly dispersed systems. Complementarily, Dinda et al. [104] employed the metal-free radical mediator 2,2,6,6-tetramethylpiperidine-1-oxyl (TEMPO) as a soluble electrocatalyst, showing that pH alone can switch product selectivity, pH 10 favoring glycolate/oxalate and pH 14 promoting C–C cleavage to formate/carbonate, while maintaining simultaneous HER at the cathode. Together, these studies illustrate that organic catalysts (both polymeric and molecular) provide a versatile, metal-lean route for controllable PET electro-upgrading, where selectivity is governed primarily by molecular redox chemistry and reaction environment rather than surface reconstruction of solid catalysts.

3.5 Hybrid catalysts

3.5.1 Multicomponent hybrid catalysts

The performance of electrocatalysts is no longer dictated solely by the intrinsic activity of individual materials; instead, it is increasingly governed by rational materials integration and interfacial engineering [74, 103]. In recent years, constructing heterojunctions, such as LDH/metal [105–107], LDH/oxide, LDH/sulfide [108], MOF/oxide, metal/carbon [23], or organic–inorganic interfaces [109, 110], by coupling two distinct catalytically active components has emerged as a powerful strategy to overcome the inherent limitations of single-phase catalysts [111]. Such heterostructures can integrate complementary functions, including efficient electron transport, optimized adsorption of reactants and intermediates, enhanced water activation, and improved structural stability, while interfacial charge redistribution accelerates reaction kinetics [12, 21]. As a result, well-designed heterojunction catalysts often deliver lower overpotentials, higher current densities, improved selectivity, and superior durability for both EGOR and HER.

Building on this general concept, a particularly influential approach has been the hybridization of precious metals with transition-metal compounds, which seeks to retain the high intrinsic activity of noble metals while minimizing their loading and simultaneously enabling bifunctional performance for coupled EGOR||HER systems [112]. In this regard, Du et al. [113]

developed a representative Pt/ γ -NiOOH/Ni foam hybrid catalyst, where sparsely dispersed Pt nanoparticles were anchored on vertically aligned γ -NiOOH nanosheets. The intimate Pt–NiOOH interface induced strong electronic metal–support interactions, thereby enhancing EG adsorption and lowering the energy barrier of the potential-determining step. As a result, this catalyst enabled selective conversion of real PET hydrolysate to glycolate with > 90% FE and selectivity at a marginal potential of 0.55 V, while simultaneously delivering efficient HER at the cathode, highlighting how rational integration of precious metals with redox-active transition-metal oxyhydroxides can substantially improve both performance and techno-economic viability (Figs. 8(a) and 8(b)).

Complementary to noble-metal hybrids, heterojunctions constructed within transition-metal compounds have also become a central design strategy in electrocatalysis. While such internal heterointerfaces can induce beneficial charge redistribution, optimize electronic structures, and provide complementary redox functions for both EGOR and HER, their effectiveness remains highly system-dependent. Poorly matched phase combinations may still suffer from interfacial instability, mismatched adsorption energetics, or blocked active sites, which can compromise performance in coupled EGOR||HER systems. A compelling demonstration of rational design was reported by Qi et al. [33], who proposed a Cr–Ni₃S₂–Ni(OH)₂/NF heterostructure for PET-derived EG oxidation. Upon activation, Ni(OH)₂ converts to Ni³⁺–OOH as the key active species. Cr and S regulate the Ni electronic structure via Ni–S–Cr bridging, stabilizing Ni³⁺ and accelerating the Ni²⁺/Ni³⁺ cycle. The intimate sulfide/hydroxide interface increases electrochemically active surface area (ECSA) and lowers charge-transfer resistance, enabling industrial-level EGOR (1.561 V at 1200 mA·cm^{−2}). When coupled with HER, the catalyst replaces OER and significantly reduces cell voltage. The EGOR||HER system delivers 1200 mA·cm^{−2} at only 1.828 V while co-producing formate and H₂, demonstrating the dual-functional advantage of this heterostructure for energy-efficient paired electrolysis (Figs. 8(c)–8(f)).

Extending this heterojunction concept to porous crystalline materials, MOF-based hybrid architecture has recently gained prominence. Beyond single-component MOFs, coupling MOFs with other catalytically active materials integrates high porosity and tunable coordination chemistry with improved electrical conductivity and water-activation capability. Importantly, such rationally engineered MOF hybrids can function as bifunctional catalysts, simultaneously driving EGOR at the anode and HER at the cathode, thereby enabling efficient paired electrolysis within a single material platform. In this context, Li et al. [31] designed a Ni–MOF@MnCo–OH hierarchical heterojunction for PET-derived EG upgrading. Ni–MOF acts as the primary EGOR-active phase due to its strong EG adsorption and Ni²⁺/Ni³⁺ redox activity, while MnCo–OH supplies abundant adsorbed hydroxyl species (OH_{ad}) via efficient water activation. The interfacial coupling improves charge transfer and intrinsic activity, enabling PET upgrading with ~ 80% FE at 300 mA·cm^{−2} (1.43 V vs. RHE). The heterostructure concept was further extended to HER using Ni–MOF@MnCo₂O_{4.5}, which shows a low overpotential (198 mV at 10 mA·cm^{−2}) and nearly 100% H₂ FE when paired with PET upgrading. This work highlights MOF-based hybrids as bifunctional platforms for integrated EGOR and HER.

3.5.2 Catalyst–support interface engineering

Depositing catalytically active materials onto conductive substrates

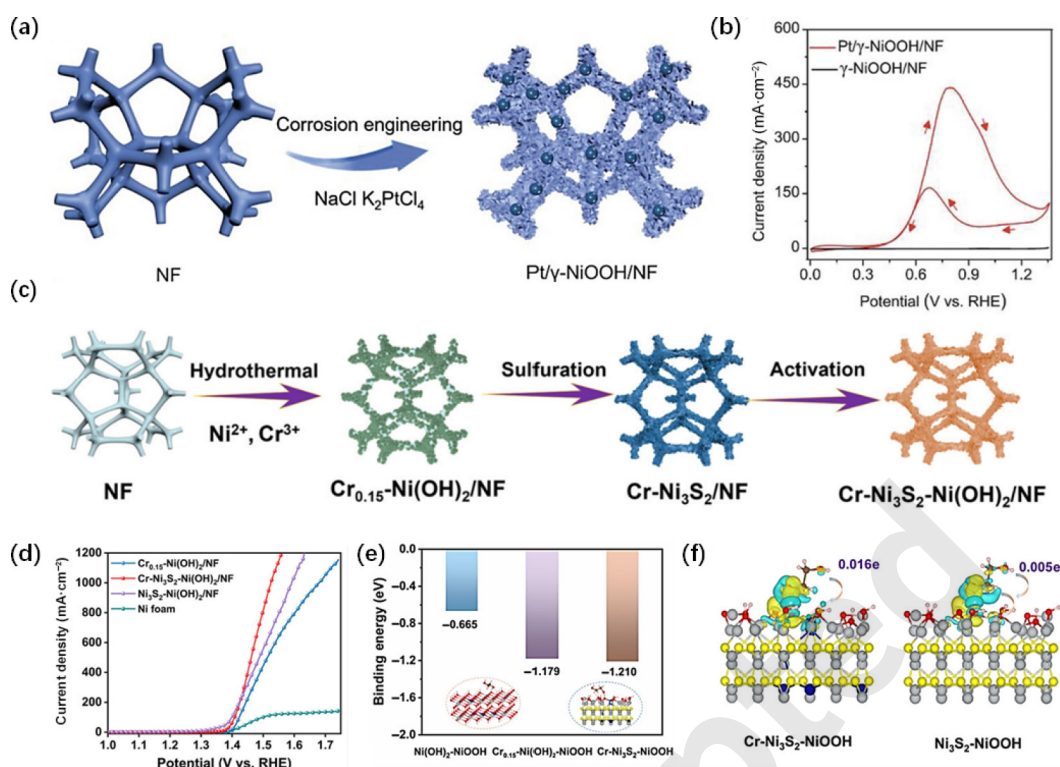


Figure 8 (a) Schematic illustration of the fabrication of Pt/γ-NiOOH/NF via a corrosion engineering strategy. (b) Cyclic voltammetry (CV) profiles of Pt/γ-NiOOH/NF and bare γ-NiOOH/NF recorded (0.3 M EG + 1.0 M KOH). Reproduced with permission from Ref. [113], © Wiley-VCH GmbH 2023. (c) Schematic illustration of the preparation of Cr-Ni₃S₂-Ni(OH)₂/NF on nickel foam. (d) Electrochemical evaluation of Cr_{0.15}-Ni(OH)₂/NF, Ni₃S₂-Ni(OH)₂/NF, and Cr-Ni₃S₂-Ni(OH)₂/NF catalysts for EGOR. (e) DFT-calculated adsorption energies of EG on Ni(OH)₂-NiOOH, Cr_{0.15}-Ni(OH)₂-NiOOH, and Cr-Ni₃S₂-NiOOH surfaces. (f) Charge density difference maps for EG* adsorption on Cr-Ni₃S₂-Ni(OH)₂/NF and Ni₃S₂-NiOOH. Reproduced with permission from Ref. [33], © Wiley-VCH GmbH 2025.

has emerged as a widely adopted strategy to enhance electrocatalytic performance for both EGOR and HER [23, 114]. In such systems, the conductive substrate primarily functions as an efficient electron-transport network and mechanical scaffold, while the loaded active phase provides the reactive sites for EG adsorption/oxidation and hydrogen evolution. Highly conductive carbon-based supports, such as carbon cloth, carbon nanotubes (CNTs), and graphene, act as effective current collectors and offer large surface areas for anchoring nanostructured catalysts, thereby mitigating particle aggregation during operation [115]. By lowering charge-transfer resistance and accelerating interfacial electron transport, these supports substantially improve reaction kinetics for both anodic EGOR and cathodic HER. Moreover, strong interfacial interactions between the active material and the substrate can stabilize dispersed active sites and, in some cases, modulate the local electronic structure and reaction microenvironment to favor desired pathways. Through rational selection of conductive substrates and optimized loading architectures, this strategy enables simultaneous enhancement of activity, selectivity, and durability in coupled EGOR||HER systems.

Zhang et al. [63] developed a PdFe alloy supported on N-doped CNTs (PdFe/N-CNTs) to create a metal/N-carbon interfacial heterojunction. Fe modulates the electronic structure of Pd while N-CNTs provide a conductive and stable support. PdFe/N-CNTs exhibit the lowest onset potential and highest current density for EGOR in 1 M KOH + 0.3 M EG, outperforming Pd/C, Fe/CNTs, and RuO₂, indicating enhanced intermediate activation at the heterointerface. The Tafel plots reveal the smallest slope (~ 54 mV·dec⁻¹), evidencing accelerated charge-transfer kinetics.

Based on this, an anion-exchange membrane (AEM) flow cell using PdFe/N-CNTs as bifunctional electrodes was constructed for paired EGOR||HER. The full-cell polarization shows that EGOR||HER requires substantially lower voltage than OER||HER, demonstrating energy-saving operation. Finally, across 100–1000 mA·cm⁻², EGOR||HER consistently operates at lower voltage, highlighting the system-level advantage of the PdFe/N-CNT heterojunction design.

Loading catalytically active materials onto MXenes to form metal-MXene interfacial heterojunctions has emerged as an effective strategy for enhancing both EGOR and HER. Owing to their metallic conductivity and high carrier density, MXenes act as efficient electron highways that lower charge-transfer resistance between the support and active phase. Moreover, abundant surface terminations (–O, –OH, and –F) provide robust anchoring sites that suppress nanoparticle agglomeration and enhance durability. Overall, MXene-based heterojunction catalysts offer a unified platform to simultaneously optimize activity, selectivity, and stability in coupled EGOR||HER systems. Yu et al. [114] demonstrated that strong interfacial coupling between CoCuO_x active species and Ti₃C₂T_x MXene significantly enhances EGOR performance during PET upcycling. CoCuO_x/MXene delivers a lower EGOR onset potential and higher current density than Co@MXene, Cu@MXene, and RuO₂. The same catalyst shows a reduced HER overpotential. The intimate CoCuO_x-MXene coupling not only provides fast electron transport pathways but also stabilizes the active metal oxide phase and optimizes the local reaction microenvironment. Benefiting from the conductive MXene support, the CoCuO_x/MXene catalyst achieved a high FE of ~ 87.6% toward FA at 1.37 V vs. RHE, while

simultaneously enabling hydrogen production at the cathode. Notably, the hybrid catalyst maintained stable operation at industrially relevant current densities of 10 mA·cm⁻², outperforming CoCuO_x without MXene support.

Table 1 summarizes the intrinsic characteristics, advantages, and practical limitations of the representative catalyst classes for coupled EGOR||HER from a materials-chemistry and engineering perspective. Noble-metal catalysts exhibit the highest intrinsic bifunctional activity and well-defined surface states, making them indispensable as mechanistic model systems and performance benchmarks. However, their high cost and limited tunability in reaction pathways fundamentally restrict large-scale deployment. Alloying or high-entropy engineering partially alleviates this issue by enabling simultaneous optimization of EGOR intermediate adsorption and hydrogen binding energetics, although the increased structural complexity introduces challenges in stability,

reproducibility, and scale-up. In contrast, oxyhydroxides, LDHs, and transition-metal oxides offer outstanding structural robustness and economic viability. Their redox-active metal centers and OH-rich surfaces are particularly favorable for high-current EG oxidation, but their relatively limited conductivity and moderate HER kinetics necessitate further interfacial or compositional regulation for fully integrated paired electrolysis. Chalcogenides, phosphides, and nitrides represent a distinct class of conductive and dynamically reconstructed catalysts, in which metallic character and rapid charge transfer enable intrinsically balanced EGOR and HER activity, making them especially suitable for symmetric or quasi-symmetric electrolyzer configurations. MOF-based and organic molecular systems, on the other hand, provide unparalleled tunability in active-site coordination environments and reaction selectivity, yet their insufficient conductivity and limited durability under industrial

Table 1 Advantages and limitations of catalyst classes for coupled EGOR||HER

Catalyst class	Typical features	Key advantages for EGOR HER	Main drawbacks	Best-fit positioning
Precious metals	Single-metal surfaces with well-defined states	High intrinsic EGOR and HER activity; clear mechanistic interpretability and benchmarking value	High cost; limited tunability of selectivity/pathways; constrained scalability	Benchmarking and mechanistic studies
Doped precious metals	Electronic-structure tuning (charge redistribution, d-band modulation)	Simultaneous optimization of EGOR intermediate adsorption and HER H ⁺ binding; reduced noble-metal loading; strong synergistic effects	Structural/compositional complexity; risk of leaching/phase segregation; reproducibility and scale-up challenges	High performance with engineering validation needed
Oxyhydroxides	Dynamic high-valence centers; OH-rich surfaces	Favorable for high-rate EGOR via redox-active sites; can promote alkaline HER by facilitating water dissociation	Often limited conductivity; cathodic stability and phase evolution require attention	High-current EGOR-oriented systems
LDHs	Tunable composition; layered OH-rich architecture	Surface reconstruction to (oxy)hydroxides enables EGOR; enhanced OH ⁻ /ion transport; multimetal synergy benefits selectivity and HER Volmer step	“True active phase” becomes complex due to reconstruction; structural retention at high current needs verification	Selectivity control and electrolyte compatibility
Oxides	Rigid lattices; multivalent redox chemistry	High structural robustness and corrosion resistance; entropy/multimetal engineering enables electronic regulation	Lower conductivity can limit high-current operation; HER often needs additional optimization	Durability-first platforms
Sulfides	High conductivity; <i>in situ</i> reconstruction common	Conductive cores enable fast charge transport; reconstructed (oxy)hydroxide shells are active for EGOR; can enhance HER kinetics	Anodic reconstruction/leaching; long-term stability and impurity tolerance must be validated	Low-resistance, high-rate devices
Selenides	More metallic character; higher polarizability than sulfides	Faster interfacial charge transfer; improved kinetics for both EGOR and HER	Concerns on Se stability, cost, and environmental/safety aspects; synthesis control	Kinetics-driven, high-rate operation
Phosphides	Metallic conductivity; facile surface reconstruction	Reconstructed (oxy)hydroxides drive EGOR; favorable H ⁺ adsorption/desorption for HER; suitable for symmetric cells	Reconstruction complicates phase/structure; possible P loss during long-term operation	Symmetric/quasi-symmetric electrolyzers
Nitrides	Metallic-like conductivity; N-induced electronic tuning	ΔG_{H^+} closer to thermoneutral and improved water dissociation; also benefits *OH-associated steps in EGOR	High-temperature synthesis; surface oxidation/passivation risks	Cost-effective bifunctional candidates
MOFs	Site/porosity designability; often needs conductivity enhancement	Tunable metal sites and ordered pores aid adsorption/transport; carbon confinement/heterointerfaces can enable bifunctionality	Intrinsically poor conductivity (without hybridization); stability at high current remains challenging	Structure–function design and mechanistic insight
Organic/molecular catalysts	Outer-sphere electron transfer; environment-controlled selectivity	Highly tunable selectivity (pH/potential window); low metal reliance	Mediator recovery and crossover/side reactions; industrial current density and durability remain challenging	Selectivity and mechanistic exploration
Heterojunctions	Interfacial synergy; division of functions	Interface-driven charge redistribution and functional complementarity (EG adsorption, water activation, conductivity, anti-poisoning)	Performance depends on interface quality; interface instability or site blocking possible	Main route toward “high performance + practicality”
Supported catalysts	Conductive scaffolds and strong interfacial coupling	Lower resistance, higher ECSA, suppressed aggregation; MXenes can act as electron highways	Support corrosion/delamination; manufacturing consistency for scale-up	Device integration and scale-up engineering

current densities confine them primarily to structure–function design and mechanistic studies. Hybrid and supported catalysts integrate the complementary functions of different components through interfacial electronic coupling and conductive scaffolds, thereby offering a viable pathway toward simultaneously achieving high activity, controllable selectivity, and practical device compatibility.

Based on these material-level descriptors, Fig. 9 provides a system-level radar comparison that visualizes the multi-objective trade-off among activity, selectivity, durability, economic viability, and scalability. The radar map clearly shows that noble-metal systems occupy the activity-dominated region but are severely constrained by cost and scale-up simplicity. Oxyhydroxide/hydroxide/oxide platforms exhibit the highest durability and economic feasibility, confirming their suitability as industrial EGOR anodes, although their bifunctional performance remains moderate. Notably, chalcogenide/phosphide/nitride catalysts display the most balanced distribution across all descriptors, indicating their unique advantage as practical bifunctional platforms for coupled EGOR||HER, particularly in symmetric electrolysis systems. MOF and organic catalysts are positioned in the selectivity-dominated region, reflecting their strong capability for reaction pathway regulation but limited large-current applicability. Hybrid catalysts occupy the region closest to the ideal multi-objective envelope, demonstrating that interfacial synergy is currently the most promising strategy for simultaneously optimizing intrinsic activity and system-level practicality. Overall, the combined analysis of Table 1 and Fig. 9 reveals that catalyst selection for coupled EGOR||HER is governed not by a single performance metric but by the balance between intrinsic activity, structural stability, economic viability, and device integration requirements. This analysis establishes a catalyst selection roadmap for practical EGOR||HER electrolyzers, where chalcogenide/phosphide/nitride catalysts provide the best system-level balance for practical EGOR||HER devices, while noble metals serve as benchmarks, oxyhydroxides/oxides are ideal industrial EGOR anodes, and hybrid systems represent the most promising high-performance design strategy.

4 Electrochemical cell design for EGOR||HER

Electrochemical cell design is a central determinant of the practical performance of EGOR||HER paired electrolysis [18]. While

catalyst development has been the primary focus of most studies, the reactor architecture ultimately governs mass transport, Ohmic resistance, product crossover, gas–liquid management, and long-term system stability [25]. In EGOR-centered systems, the cell must simultaneously accommodate liquid-phase organic reactants and oxidation products at the anode and gas-phase HER at the cathode [116]. More importantly, the performance of paired electrolysis cannot be assessed solely in terms of cell voltage or FE; product separation efficiency emerges as an equally critical system-level metric, as it directly affects downstream purification, product purity, and overall techno-economic feasibility.

4.1 H-type reactor

Currently, H-type reactors for EGOR||HER can be broadly categorized into two configurations: Single-cell systems and dual-cell systems separated by an ion-exchange membrane. As illustrated in Fig. 10, the single-cell configuration integrates both anodic and cathodic reactions within the same electrolyte, leading to a simplified device structure, shortened ionic transport distance, and reduced internal resistance [90]. These features are advantageous for achieving high current densities and lowering capital and operational costs, making this configuration particularly attractive for scale-up and symmetric electrolyzer designs [75]. However, the lack of spatial separation inevitably results in the crossover of liquid-phase oxidation products and hydrogen gas, which may induce parasitic reactions, decrease FE, and increase the complexity of downstream separation [17].

In contrast, the dual-cell configuration spatially isolates the anodic and cathodic environments using an ion-exchange membrane, enabling independent optimization of electrolyte composition, pH, and catalyst stability windows for EGOR and HER [38, 51, 117]. This architecture effectively suppresses product crossover and allows efficient collection of liquid organic products and high-purity hydrogen, thereby significantly improving product separation efficiency [63]. Such a configuration is therefore essential for mechanistic studies, accurate intrinsic activity evaluation, and processes targeting high-value chemicals [92, 113]. The trade-off lies in the increased Ohmic resistance, additional membrane cost, and more complex system integration.

This comparison highlights that reactor configuration, together with catalyst platform selection, defines the practical performance boundary of EGOR||HER systems. Single-cell electrolyzers are

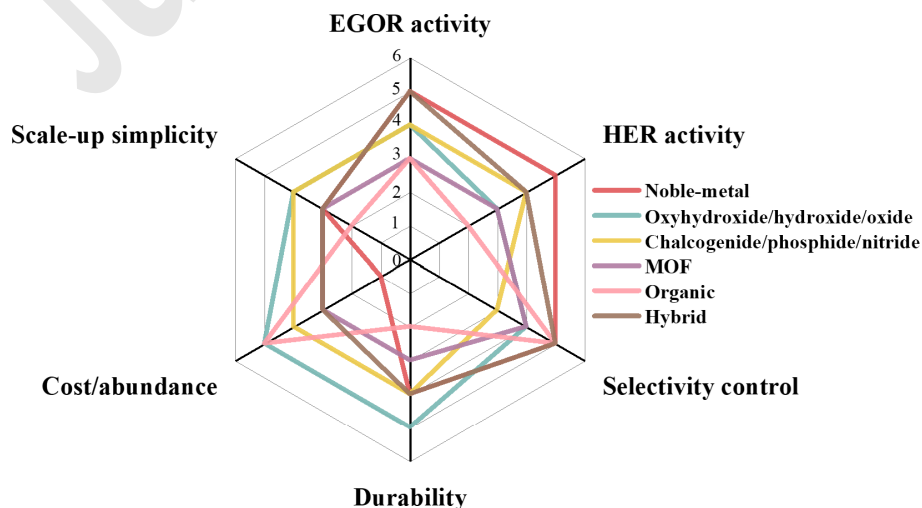


Figure 9 Radar comparison of representative catalyst platforms for coupled EGOR||HER, including noble metals, oxyhydroxide/hydroxide/oxide, chalcogenide/phosphide/nitride, MOF, organic, and hybrid catalysts.

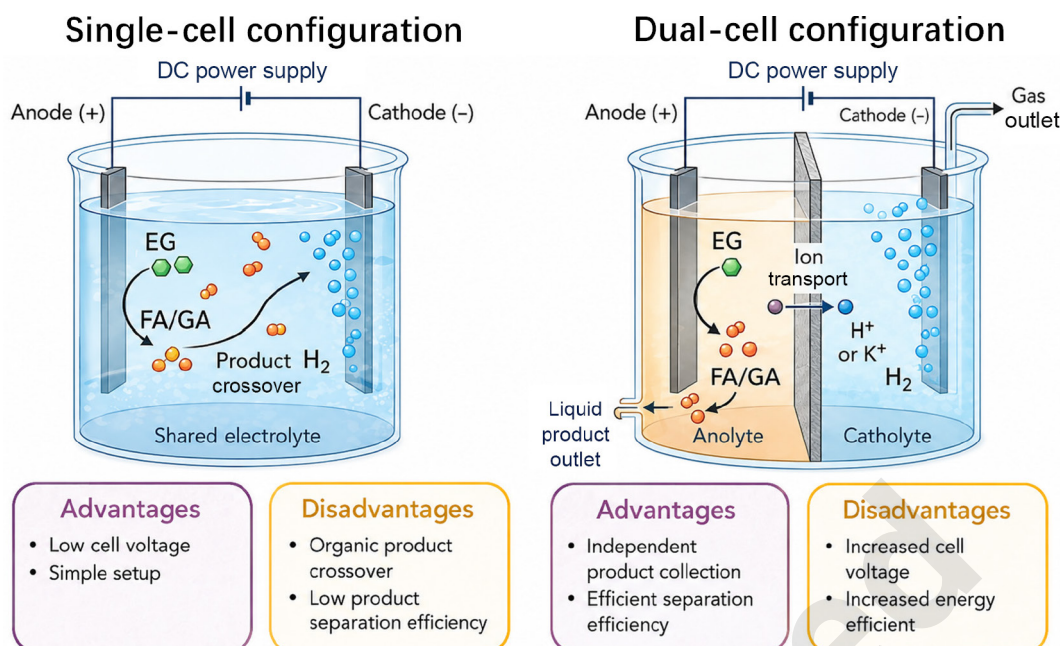


Figure 10 Comparison of single-cell and dual-cell configurations for EGOR||HER paired electrolysis.

more suitable for cost-sensitive, high-current, and scale-up-oriented applications, whereas dual-cell systems are indispensable when product purity, independent reaction optimization, and techno-economic viability are prioritized.

4.1.1 Single cell

Single-cell configurations, in which anodic EGOR and cathodic HER occur in the same electrolyte without physical separation, represent the simplest and most widely used platform for laboratory studies [42, 85]. These systems are attractive due to their low complexity, low internal resistance, and ease of assembly. In such configurations, both electrodes share a common electrolyte, enabling minimal ionic path length and reduced Ohmic losses [91]. As a result, single-cell setups often exhibit lower operating cell voltages compared with separated systems, which is beneficial for evaluating the intrinsic energy advantages of EGOR. Wang et al. [46] reported a membrane-free single-cell electrolyzer for coupling PET hydrolysate oxidation with hydrogen production. As shown in Fig. 11(a), the system uses PET hydrolysate as a shared electrolyte, where the cathode drives HER while the anode converts EG to formate. Figure 11(b) shows that within 1.5–1.9 V, the device maintains high selectivity for both products (FE for H₂ (FE_{H_2}) = 95%–100%, $FE_{formate}$ = 80%–95%), demonstrating that a single-chamber design can enable energy-efficient co-electrolysis with excellent product selectivity. As another example of a membrane-free single-cell electrolyzer, Wang et al. [118] employed identical N-Ni₃P-NiMoO₄/NF electrodes for EGOR||HER hydrolysate oxidation (Fig. 11(c)), allowing both half-reactions to occur in the same PET electrolyte without an ion-exchange membrane. Figure 11(d) shows that replacing OER with EGOR lowers the cell voltage by ~194 mV at 100 mA·cm⁻² compared with 1 M KOH water electrolysis, evidencing significant energy savings. Meanwhile, Figs. 11(e) and 11(f) demonstrate high product selectivity in this single chamber, with $FE_{formate}$ = 80%–95% and FE_{H_2} = ~100% over 1.4–1.8 V, confirming efficient simultaneous production of formate and hydrogen.

However, the lack of physical separation introduces several

critical limitations that restrict the relevance of single-cell configurations for practical applications. First, product crossover is unavoidable. Oxidation products, such as formate, glycolate, or intermediate species, can diffuse to the cathode, where they may be reduced or undergo parasitic reactions. This not only decreases FE but can also complicate product analysis and purification. Second, gas evolution and liquid-phase organic reactions compete within the same electrolyte volume. Hydrogen bubbles generated at the cathode can disturb hydrodynamics and influence mass transport of EG and oxidation products near the anode. This coupling can obscure intrinsic reaction kinetics and lead to apparent performance that is highly sensitive to stirring, electrode spacing, and cell geometry. As a result, data obtained in single-cell systems are often difficult to scale or compare across different studies.

4.1.2 Dual cell

Dual-cell configurations spatially separate the anodic EGOR and cathodic HER using a membrane or separator, thereby enabling independent control of the anolyte and catholyte and separate collection of liquid organic products and hydrogen gas [12, 14]. Such configurations can be implemented in H-type cells, flow electrolyzers, or membrane-electrode assemblies (MEAs) [94, 111].

This architecture represents a critical step toward practical implementation of EGOR||HER paired electrolysis, as it directly addresses the separation limitations of single-cell systems [29]. The anolyte composition, pH, and additives can be tailored to maximize EGOR activity and selectivity toward FA or GA, while the catholyte can be independently optimized for efficient HER and gas handling [119]. The key advantage of dual-cell configurations is their significantly improved product separation efficiency. Physical separation prevents direct mixing of anodic liquid products and cathodic species, enabling higher effective recovery of FA and GA and reducing the risk of parasitic cathodic reactions [120]. From a process perspective, this improved separation efficiency directly reduces downstream purification requirements and enhances the overall feasibility of valorizing liquid oxidation products. However, this benefit is accompanied

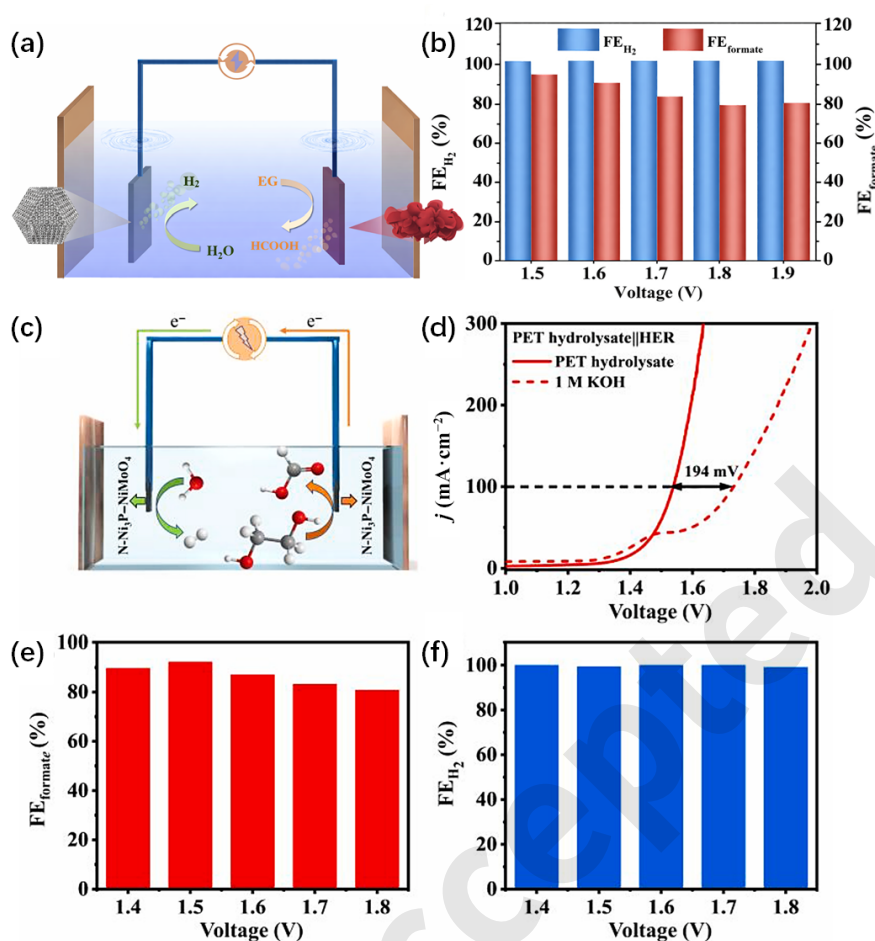


Figure 11 Paired electrolysis in a single-cell, membrane-free cell: (a) schematic of the EGOR||HER paired electrolysis using bifunctional ordered macroporous superstructure Ni-modified CoP (OMS-Ni-CoP) electrodes in a single electrolyzer. (b) FE for FA in the OMS-Ni-CoP||OMS-Ni-CoP cell in 1 M KOH. Reproduced with permission from Ref. [46], © Elsevier B.V. 2022. (c) Schematic of the membrane-free EGOR||HER based on N-Ni₃P-NiMoO₄/NF. (d) LSV curves of the N-Ni₃P-NiMoO₄/NF||N-Ni₃P-NiMoO₄/NF cell in 1 M KOH with and without PET hydrolysate. ((e) and (f) FE for formate and H₂ in the same single-cell system. Reproduced with permission from Ref. [118], © Elsevier Ltd. 2023.

by increased internal resistance due to the presence of a membrane or separator. The resulting Ohmic losses lead to higher operating cell voltages, which can erode part of the thermodynamic advantage provided by EGOR relative to OER. This introduces a fundamental trade-off between energy efficiency and separation efficiency. Architectures that maximize product separation may incur higher voltage penalties, whereas low-resistance designs may compromise product retention. In addition, ion transport across the separator can induce local pH and concentration gradients, which can influence EGOR kinetics and pathway selectivity. Such effects may shift the balance between C1 and C2 pathways and alter long-term performance.

Separators and membranes are central components in dual-cell EGOR||HER systems, as they directly govern not only ionic conductivity and cell voltage, but also product separation efficiency and effective product recovery [108, 121]. Membranes function as key system-level regulators that determine how efficiently liquid oxidation products and H₂ gas can be spatially separated and collected. AEM [9, 50, 52, 105, 122] and proton exchange membrane (PEM) [123] are the most employed membrane types in EGOR||HER systems.

AEMs are frequently favored because they facilitate hydroxide ion transport, which supports both EGOR and HER under alkaline conditions. Jin et al. [107] constructed an AEM-separated flow cell to demonstrate practical operation of EGOR. In Fig. 12(a),

the cell adopts an AEM to spatially separate the anodic Pd/Ni(OH)₂/NF catalyst from the cathodic Pt/C electrode, allowing OH⁻ transport. The AEM configuration enables independent control of the anolyte (EG + KOH) and catholyte (KOH), which is essential for maintaining high EGOR activity at the anode while sustaining efficient HER at the cathode. In Fig. 12(b), the AEM-based flow cell exhibits lower cell voltage in 1 M EG + 1 M KOH compared with 1 M KOH alone, confirming that replacing water oxidation with EGOR substantially reduces the energetic demand of the overall process in an AEM system. However, many AEMs also permit partial transport of organic anions, such as formate and glycolate. This organic crossover directly reduces product separation efficiency by allowing anodic products to migrate into the cathode cell, lowering effective product recovery, decreasing apparent FE, and increasing the burden on downstream liquid-phase separation and purification. Membrane crossover in EGOR||HER systems is strongly affected by AEM properties, such as ion-exchange capacity, thickness, water uptake, hydration degree, and ion-channel structure. Although high ion-exchange capacity and strong hydration improve OH⁻ conductivity, they may also enlarge transport channels and promote the crossover of organic anions, such as FA and GA. Increasing membrane thickness can reduce crossover, but it also raises Ohmic resistance. Therefore, future AEM design should focus on membranes with high OH⁻ conductivity, low

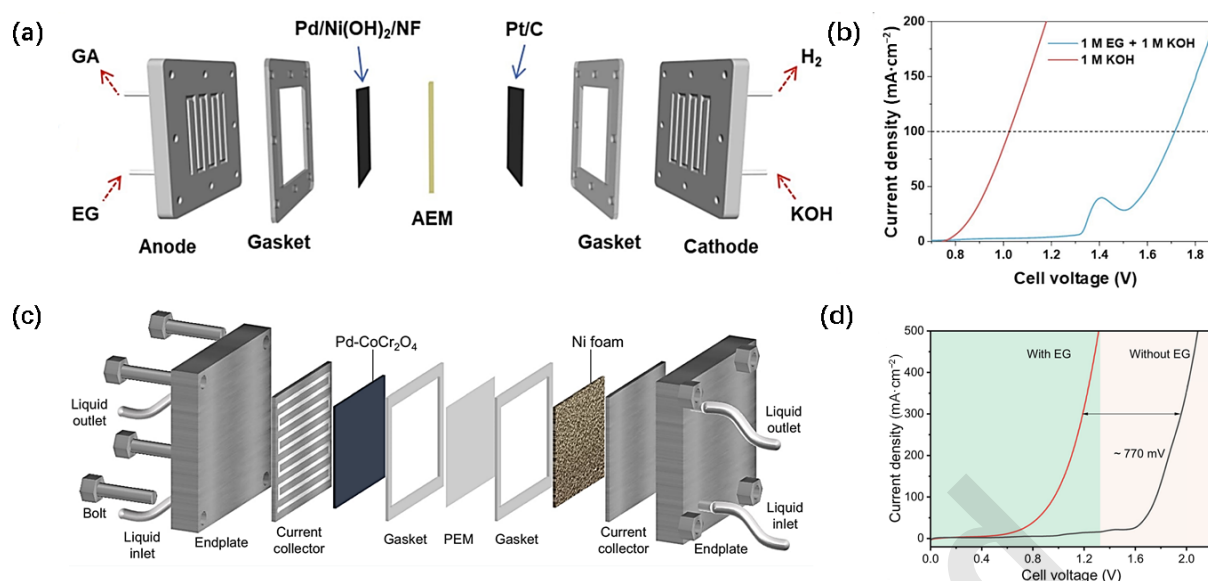


Figure 12 Schematic and electrochemical performance of Dual-cell configurations for EGOR and H₂ production: (a) schematic representation of a mobile electrolyzer based on an AEM. (b) LSV curves for EGOR in the AEM-separated flow cell. Reproduced with permission from Ref. [107], © Elsevier Inc. 2025. (c) Schematic diagram of a membrane electrode flow cell employing a PEM to separate the anode and cathode for the continuous electrosynthesis of GA from EG coupled with H₂ generation. (d) LSV curves of the PEM-based flow-cell system with and without EG at a high flow rate of 10 mL·min⁻¹. Reproduced with permission from Ref. [124], © Wang, Y. X. et al. 2025.

organic-anion permeability, controlled swelling, and selective ion-channel structures.

In contrast, PEMs can more effectively suppress the transport of anionic organic species, thereby improving anodic product retention and enhancing intrinsic product separation efficiency. However, this benefit is often accompanied by higher membrane resistance in alkaline environments, leading to increased Ohmic losses and higher operating cell voltages. In addition, cation-exchange membranes (CEMs) can alter local pH near the anode, which may negatively affect EGOR kinetics and shift pathway selectivity between C1 and C2 products. Wang et al. [124] developed a flow cell to couple EGOR with HER. In this design, a Pd-CoCr₂O₄ anode and a Ni-foam cathode are separated by a Nafion PEM, which prevents product crossover while enabling proton transport. The sandwich structure of current collectors, gaskets, and endplates supports stable operation under continuous flow (Fig. 12(c)). As shown in Fig. 12(d), replacing water oxidation with EGOR significantly reduces the required cell voltage at the same current density. This ~770 mV voltage drop demonstrates that the PEM-based configuration enables energy-efficient, simultaneous GA electrosynthesis and H₂ production.

Beyond conventional ion-exchange membranes, porous separators and size- or charge-selective membranes represent underexplored opportunities for improving separation performance in EGOR||HER systems. By tailoring pore size, surface chemistry, and functional groups, such separators could selectively hinder organic product transport while maintaining high ionic conductivity. Hybrid porous-ion-exchange architectures and surface-functionalized membranes offer promising pathways to decouple product separation efficiency from Ohmic resistance, enabling simultaneous improvement of energy efficiency and product purity.

Critically, most membranes currently adopted in EGOR studies are inherited from water electrolysis technologies and are not optimized for liquid organic product management. From an innovation standpoint, the development of EGOR-specific membranes that explicitly target high liquid product retention,

resistance to organic fouling, and chemical stability in organic-rich environments represents a major opportunity. Such membranes could substantially improve product recovery, reduce downstream purification burdens, and enhance the overall techno-economic viability of EGOR||HER paired electrolysis.

Table 2 summarizes the EGOR||HER paired electrolysis systems from the perspectives of catalyst composition and reactor configuration. In single-cell configurations, most systems operate at relatively low current densities (typically 10–310 mA·cm⁻²) and cell voltages of 1.3–1.7 V, although several advanced catalysts, such as Ir-PdPtAuNiCu HEAN and Au/Ni(OH)₂, enable high current densities with reduced cell voltages. In contrast, dual-cell electrolyzers based on AEM and PEM architectures allow independent optimization of anodic EGOR and cathodic HER, leading to significantly enhanced current densities (up to > 1000 mA·cm⁻²) and more stable hydrogen production with near-unity FE. From the catalytic perspective, most high-performance systems exhibit high selectivity toward formate or glycolate with Faradaic efficiencies above 90%, indicating that the product distribution is primarily governed by the anodic catalyst rather than the reactor type.

4.2 Flow-cell reactor

In the electrochemical conversion of ethylene glycol, flow-cell reactors have been increasingly used to overcome the mass-transfer limitations of conventional H-type or static cells [128]. More recently, this reactor configuration has also been introduced into EGOR||HER coupled systems, where continuous electrolyte circulation can enhance reactant supply, accelerate product removal, support higher current densities, and provide a more practical platform for evaluating coupled electrolysis [129]. Like the compartmental separation in H-type reactors, EGOR||HER flow-cell reactors can be classified into membrane-free and membrane-separated systems depending on whether the anodic EGOR and cathodic HER regions are physically isolated (Fig. 13) [130, 131].

Table 2 Comparison of reported EGOR||HER paired electrolysis systems in terms of both catalyst design and reactor configuration

Anode	Electrolyte	Voltage (V)	j (mA·cm ⁻²)	Product _{anode}	FE _{anode} (%)	Cathode	Product _{cathode}	FE _{cathode} (%)	Reactor	References
Re _{0.5} Co _{0.5} O _{2.42}	1 M KOH + EG	1.06	10	FA	90.9	Re _{0.5} Co _{0.5} O _{2.42}	H ₂	—	Single cell	[20]
m-HEA	1 M KOH + EG	1.6	250	GA	97	m-HEA	H ₂	~ 100	Single cell	[42]
Ni-Co ₉ S ₈ NSAs/NF	1 M KOH + EG	1.5	~ 140	FA	~ 92	Pt wire	H ₂	~ 100	Single cell	[18]
CoFeNi	1 M KOH + 0.3 M EG	1.52	100	FA	90.2–80.3	Pt-CoFeNi	H ₂	~100	Single cell	[65]
PdNi	1 M KOH + 0.1 M EG	1.90	100	GA	60	CuCoP	H ₂	—	Single cell	[57]
Ir-PdPtAuNiCu HEAN	1 M NaOH + 0.5 M EG	0.724	300	GA	88.8	PdPtAuNiCu HEAN	H ₂	—	Membrane-free flow cell	[41]
Au/Ni(OH) ₂	3 M KOH + 1 M EG	1.4	~ 310	GA	96	Ni foam	H ₂	~ 100	Single cell	[112]
Pd-CuCo ₂ O ₄	1 M NaOH + 0.5 M NaCl + 1 M EG	0.87	100	GA	96.1	CoP	H ₂	~ 100	Single cell	[14]
CuO/Cu	1 M KOH + EG	1.45	10	FA	88	Pt gauze	H ₂	~ 100	Single cell	[116]
CoCuO _x @MXene	1 M KOH + 0.3 M EG	1.65	100	FA	~ 90	CoCuO _x @MXene	H ₂	~ 100	Single cell	[114]
Ni-Fe ₃ Se ₄	1 M KOH + EG	1.66	100	FA	89	Ni-Fe ₃ Se ₄	H ₂	—	Single cell	[86]
CoP	1 M KOH + 0.5 M EG	1.39	100	FA	~ 92	CoP	H ₂	~ 100	Single cell	[125]
OMS-Ni ₁ -CoP	1 M KOH + 0.5 M EG	1.52	10	FA	—	OMS-Ni ₁ -CoP	H ₂	—	Single cell	[46]
Ni ₂ P	1 M KOH + EG	1.46	50	FA	~ 88	Ni ₂ P	H ₂	~ 100	Single cell	[126]
Pd-NiTe	1 M KOH + EG	1.6	10	FA	95.6	Pd-NiTe	H ₂	98.6	Dual cell (MEA)	[29]
Ni _{150s}	1 M KOH + 0.1 M PET	1.5–1.8	100	FA	~ 90	Ni _{150s}	H ₂	~ 100	Dual cell (AEM)	[122]
PdFe/N-CNT	1 M KOH + EG	1.68	500	FA	87	PdFe/N-CNT	H ₂	~ 100	Dual cell (MEA)	[63]
Ru _{SA} /NC/Ni(OH) ₂	1 M KOH + EG	1.6	> 1000	FA	94.3	Pt	H ₂	—	Dual cell (PEM)	[6]
NiFe-LDH	1 M KOH + EG	1.41	100	FA	94.3	Pt	H ₂	—	Dual cell (MEA)	[121]
Pt/γ-NiOOH	1 M KOH + EG	0.55	—	GA	95	Pt/γ-NiOOH	H ₂	~ 100	Dual cell (MEA)	[113]
Ni ²⁺ -CoOOH	1 M KOH + 0.3 M EG	2.16	1000	FA	> 90	MoNi ₄ @MoO ₂	H ₂	~ 100	Dual cell (MEA)	[70]
CoV-LDH	1 M KOH + EG	1.63	50	FA	86.61	VP-CoP	H ₂	—	Dual cell (AEM)	[71]
Pd/Ni(OH) ₂	1 M KOH + EG	1.02	100	GA	94.2	Pt/C	H ₂	—	Dual cell (AEM)	[107]
Ni(OH) ₂ -Ni ₃ S ₂	1 M KOH + 0.5 M EG	—	70	FA	89	Ni(OH) ₂ -Ni ₃ S ₂	H ₂	—	Dual cell (AEM)	[108]
B ₂ Co-NiS	1 M KOH + EG	1.491	100	FA	> 93	B ₂ Co-NiS	H ₂	~100	Dual cell (MEA)	[127]
Pd/Ni ₃ S ₂	1 M KOH + EG	1.70	50	FA	95.6	Pt/Ti felt	H ₂	—	MEA flow cell	[84]
NiSe ₂ /GC	1 M KOH + EG	1.395	50	FA	> 80	Pt wire	H ₂	~ 100	Dual cell (PEM)	[87]
Co ₂ P-Ni ₂ P/CC	1 M KOH + EG	1.473	100	GA	> 90	Co ₂ P-Ni ₂ P/CC	H ₂	> 95	Dual cell (AEM)	[9]
N-Ni ₃ P-NiMoO ₄	1 M KOH + EG	1.53	100	FA	92.2	N-Ni ₃ P-NiMoO ₄	H ₂	~ 100	Single cell	[118]
Co-CeF ₃ @CC	1 M KOH + 0.5 M EG	1.69	10	FA	93.5	Co-CeF ₃ @CC	H ₂	—	Dual cell (MEA)	[94]

4.2.1 Membrane-free

Membrane-free flow cells provide a simplified reactor configuration for EGOR||HER coupling by eliminating the need for an ion-exchange membrane [132]. This design reduces cell resistance and simplifies reactor assembly, but the absence of a membrane inevitably leads to mixing between anodic and cathodic products. Guo et al. [130] constructed a membrane-free EGOR||HER flow cell using N-PdRh as both the anodic EGOR and cathodic HER catalyst. The bifunctional system was assembled in a Gaoss Union flow cell, where 3.0 M KOH with or without 1.0 M EG served as the electrolyte. This membrane-free

configuration achieved 100 mA·cm⁻² at only 0.85 V, with GA selectivity above 97.6% and an H₂ production energy consumption of 2.52 kWh·m⁻³. Shi et al. [129] studied a membrane-free flow-cell system for paired EGOR||HER over Pd/NiMoO₄/NF. As shown in Fig. 14(a), Pd/NiMoO₄/NF and Ni foam were integrated as the anodic EGOR and cathodic HER electrodes, respectively, without using an ion-exchange membrane. Compared with the conventional single-cell setup, the flow cell delivered much higher current densities over the whole voltage range, reaching over 200 mA·cm⁻² at 1.6 V (Fig. 14(b)). However, because anodic and cathodic environments are not physically separated, membrane-free flow cells require highly

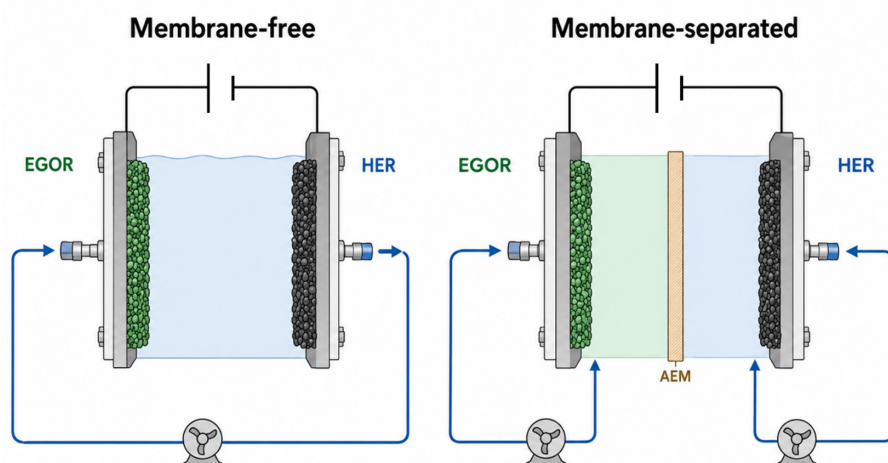


Figure 13 Schematic comparison of membrane-free and membrane-separated flow-cell reactors for EGOR||HER coupling.

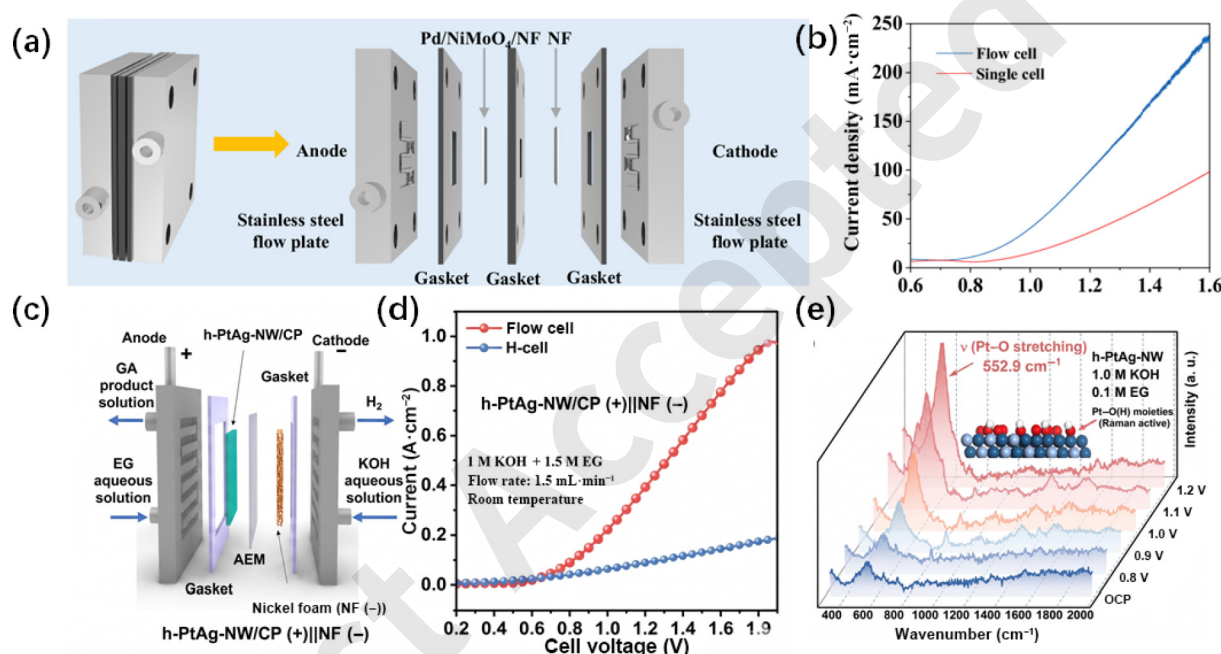


Figure 14 Flow-cell reactors for EGOR-assisted HER: (a) membrane-free flow-cell configuration using Pd/NiMoO₄/NF as the anode. (b) Current-density comparison between the flow cell and single cell. Reproduced with permission from Ref. [129], © The Authors et al. 2024. (c) AEM-separated EGOR||HER flow cell using h-PtAg-NW/CP as the anode and Ni foam as the cathode. (d) Polarization curves showing enhanced current output in the flow cell. (e) *Operando* Raman spectra revealing Pt-O(H) formation on h-PtAg-NW during EGOR. Reproduced with permission from Ref. [133], © Li, Y. H. et al. 2025.

selective bifunctional catalysts and carefully controlled electrolyte conditions to suppress undesired side reactions.

4.2.2 Membrane-separated

In contrast, membrane-separated flow-cell systems introduce an ion-exchange membrane between the anodic EGOR and cathodic HER compartments [132]. This configuration can physically separate oxidation and reduction environments, suppress product crossover, and allow independent optimization of the anolyte and catholyte compositions. Although the membrane increases device complexity and may introduce additional Ohmic resistance, it provides a more controlled platform for evaluating EGOR||HER coupling under high-current-density and long-term operating conditions. Liu et al. [131] studied an Ampere-level EGOR||HER coupled flow-cell system based on a NiFe-single-crystalline Prussian blue analog (NiFe-sc-PBA)/NF anode. The device was constructed as a membrane-electrode-assembly electrolyzer, in

which NiFe-sc-PBA/NF served as the EGOR anode and commercial 40 wt.% Pt/C-coated gas diffusion layer (GDL) served as the HER cathode. A Sustainment X37-FA anion-exchange membrane separated the two electrodes. During operation, 1 M KOH with 1 M EG was fed to the anode and 1 M KOH was fed to the cathode using peristaltic pumps at 15 rpm. The EGOR||HER system achieved stable operation at 1.0–1.5 A·cm⁻² for over 500 h, maintaining low cell voltages of 2.02–2.13 V and $FE_{FA} > 92\%$, demonstrating the feasibility of MEA flow cells for high-current-density EGOR-assisted hydrogen production. Li et al. [133] studied an AEM-based flow-cell electrolyzer for EGOR||HER coupling. The AEM-separated flow-cell configuration provides a more practical platform than conventional H-cells for EGOR||HER coupling. In the flow cell, hollowed PtAg nanowire on carbon paper (h-PtAg-NW/CP) was used as the EGOR anode, and Ni foam served as the HER cathode, with an AEM separating the anodic and cathodic compartments. Continuous electrolyte

circulation enhanced EG supply and product removal, thereby reducing mass-transfer limitations that are commonly observed in H-type cells. As shown by the polarization curves, the flow cell delivered much higher current density than the H-cell at the same cell voltage, reaching nearly $1.0 \text{ A}\cdot\text{cm}^{-2}$ at high voltage, whereas the H-cell remained below $\sim 0.2 \text{ A}\cdot\text{cm}^{-2}$. *Operando* Raman spectra further revealed the formation of surface Pt-O(H) species on h-PtAg-NW, as indicated by the Pt-O stretching band at 552.9 cm^{-1} . These surface oxygenated species facilitated selective EG oxidation toward glycolate while suppressing C-C bond cleavage, demonstrating the combined importance of catalyst surface chemistry and flow-cell reactor design for efficient EGOR-assisted hydrogen production (Figs. 14(c)–14(e)).

Computational fluid dynamics (CFD) provides a useful reactor-level tool for understanding mass transport in EGOR-based flow cells. Unlike conventional electrochemical measurements, which mainly report the overall current, voltage, and product distribution, CFD can visualize how the electrolyte flows through the reactor and how the EG concentration changes near the electrode surface. This is particularly important for high-current-density EGOR||HER operation, because rapid EG consumption at the anode can cause local EG depletion, concentration polarization, and even a shift from EGOR to OER, leading to voltage increase and reduced energy efficiency. Li et al. [132] applied CFD simulation to a membrane-free flow-cell electrolyzer for continuous EGOR||HER. As shown in Fig. 15, the reactor was

constructed with a membrane-free flow-cell configuration, in which nanoporous PdCu (n-PdCu) catalyst loaded on a gas diffusion electrode (n-PdCu/GDE) was used as the anodic EGOR electrode and paired with a cathodic HER electrode. The CFD results show that electrolyte flow rate strongly affects EG distribution on the anode surface. At low flow rates, uneven EG concentration and local depletion are observed, whereas increasing the flow rate to $50\text{--}100 \text{ mL}\cdot\text{min}^{-1}$ produces a more uniform EG concentration field. This improved reactant supply explains the more stable cell voltage under flowing conditions, while the simulated flow field and EG streamlines further reveal how forced convection enhances EG delivery to the anode surface and supports continuous high-rate operation.

According to the discussion above, reactor configuration strongly affects the practical performance of EGOR||HER coupling. Single-cell or membrane-free cells have simple structures, low resistance, and low cost, making them suitable for preliminary catalyst testing and proof-of-concept studies (Table 3). However, because EGOR and HER occur in the same electrolyte, product crossover and mixed liquid/gas products make downstream separation difficult. Dual-cell or membrane-separated cells can physically separate the EGOR anode and HER cathode, allowing independent control of the anolyte and catholyte and improving product recovery. Their main drawbacks are higher Ohmic resistance, membrane cost, and possible membrane fouling or organic-product crossover.

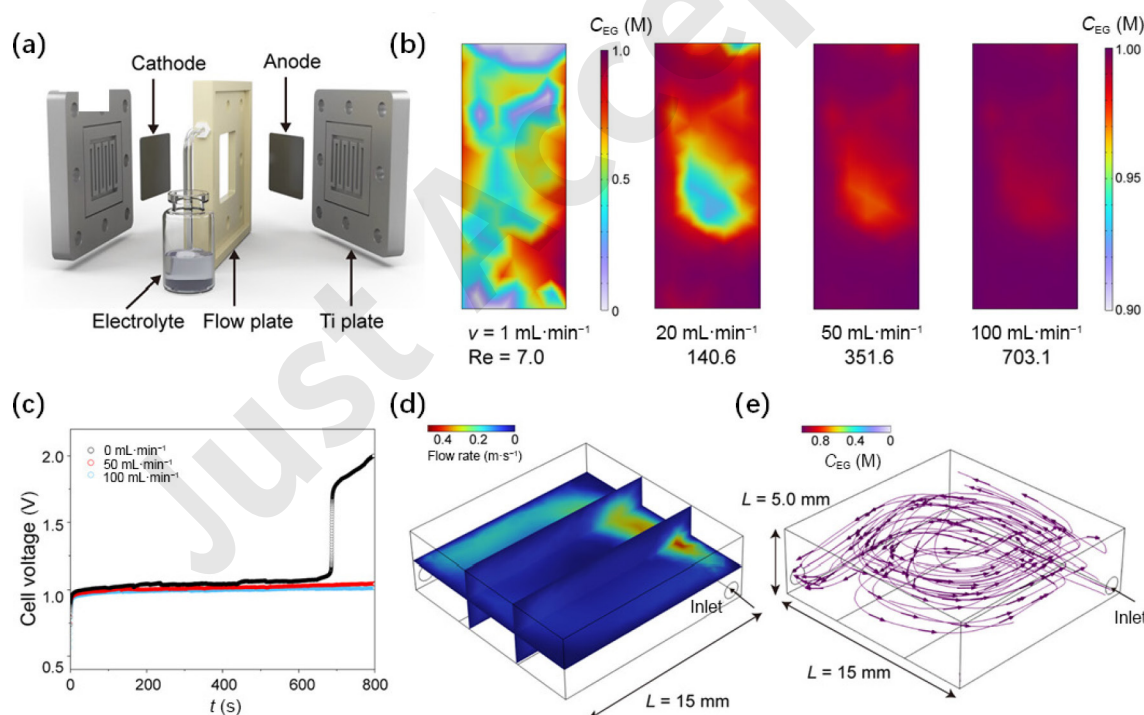


Figure 15 CFD-guided flow-cell design for EGOR||HER: (a) schematic of the membrane-free flow-cell reactor. (b) CFD-simulated EG concentration distribution on the n-PdCu/GDE anode surface at different flow rates. (c) Flow-rate-dependent chronopotentiometry at $200 \text{ mA}\cdot\text{cm}^{-2}$. ((d) and (e)) Simulated flow-field pattern and EG streamlines at $100 \text{ mL}\cdot\text{min}^{-1}$ and $200 \text{ mA}\cdot\text{cm}^{-2}$. Reproduced with permission from Ref. [132], © Li, Y. H. et al. 2026.

Table 3 Comparison of reactor configurations for EGOR||HER coupling

Reactor type	Advantages	Limitations
Single-cell static H-type reactor	Low resistance; simple structure; low cost	Product crossover; difficult separation
Dual-cell static H-type reactor	Better product separation; independent anolyte/catholyte control	Higher resistance; membrane cost and fouling
Membrane-free flow cell	Improved mass transport; continuous operation; low resistance	Requires highly selective catalysts; product mixing
Membrane-separated flow cell	High current density; better product recovery; long-term operation	More complex design; membrane stability issues

Compared with static cells, flow-cell reactors are more relevant for practical operation because continuous electrolyte circulation improves EG supply, product removal, and high-current-density performance. Membrane-free flow cells offer low resistance and simple assembly, but product mixing remains problematic. Membrane-separated flow cells or MEA reactors are more promising for practical EGOR||HER systems because they combine improved mass transport with better product separation, although membrane stability and device complexity still need improvement.

Overall, H-type and simple single-cell systems are suitable for catalyst screening, while membrane-separated flow-cell/MEA reactors should be prioritized for scale-up-oriented studies. Future reactor design should focus on reducing membrane resistance, suppressing product crossover, improving gas-liquid management, optimizing flow-field design, and validating long-term operation.

5 Challenges and future perspectives

EGOR coupled with HER has attracted increasing attention as a strategy for plastic valorization and green H_2 production. However, translating laboratory demonstrations into practical technologies still faces several challenges related to product selectivity, separation, feedstock complexity, catalyst stability, and reactor scale-up. These issues are interconnected. For example, improving product selectivity may increase downstream separation requirements, while membrane-free operation reduces electrical resistance but complicates product recovery. In this section, we summarize the key challenges and outline future research directions needed to advance this field toward practical application (Fig. 16).

5.1 C1/C2 product selectivity

Controlling selectivity between C1 products and C2 products remains a major scientific challenge. Small changes in potential, local pH, or catalysts can shift the dominant reaction pathway, leading to selectivity drift and poor reproducibility.

To improve selectivity control, future work should focus on catalyst design and reaction pathway understanding. Catalysts with tunable oxygen affinity and surface hydroxyl coverage, such as heterostructures or doped oxides, could help stabilize desired intermediates and suppress unwanted C-C bond cleavage when targeting C2 products. At the same time, *operando* characterization (e.g., Raman, IR, and online product analysis) is needed to directly observe intermediates under working conditions and identify stable selectivity windows. Reactor design also matters, using flow-through electrodes and optimized mass transport, can reduce local concentration gradients and prevent over-oxidation. Finally, selectivity should be reported consistently using FE, carbon balance, and product distribution at similar conversion levels to enable fair comparison across studies.

5.2 Product separation

Downstream product separation is a major bottleneck in PET electro-recycling. The anolyte typically contains FA or GA, excess KOH, terephthalate salts, and minor organics, while H_2 is generated at the cathode. In membrane-free single-chamber systems, the absence of a separator lowers electrical resistance but mixes products, making purification more difficult. In two-chamber systems, membranes help separate products but introduce additional resistance, membrane fouling, and higher cost. Moreover, converting alkali FA or GA into free acids usually requires acidification, which produces large amounts of waste



Figure 16 Schematic overview of the major challenges and future perspectives for coupled EGOR||HER systems.

salts. When product concentration is low, conventional distillation becomes energy-intensive, and crystallization is often limited by co-precipitation with terephthalate.

Future process development should integrate electrolysis with separation instead of treating them separately. Promising approaches include *in situ* product extraction, such as electrodialysis for selective ion removal or bipolar membranes that generate acid and base streams without excessive neutralization. For membrane-free reactors, better gas-liquid management, including rapid bubble removal and gas separators, can improve H₂ collection and prevent channel blockage. For membrane-based systems, more fouling-resistant membranes and operation protocols compatible with complex hydrolysates are needed.

5.3 Impurity tolerance

Real PET waste contains impurities, such as dyes, plasticizers, adhesives, food residues, and trace metals, which are absent in model systems. These impurities can block active sites, participate in side reactions that lower FE, and cause precipitation or fouling that limits mass transport. In addition, hydrolysis of real PET may carry over colloids or microplastics that clog porous electrodes and flow channels. As a result, performance measured with pure EG or ideal hydrolysate often overestimates stability in real wastewater.

To improve impurity tolerance, a key priority is to establish standardized and realistic experimental conditions that include representative impurities and solids so that catalyst performance can be evaluated under practical conditions. Catalyst development should target anti-fouling surfaces and materials that maintain activity in the presence of contaminants. Low-cost pretreatment steps, such as filtration, sedimentation, or electrocoagulation, should also be explored as practical ways to improve stability without extensive purification.

5.4 Catalyst durability

Long-term stability at high current densities remains a critical barrier. In alkaline anodic environments, many transition-metal catalysts undergo surface reconstruction, which can either improve activity or cause dissolution, phase segregation, or loss of conductivity. Meanwhile, organic species, such as terephthalate, can strongly adsorb on the surface and cause catalyst poisoning.

Improving durability requires a better understanding of failure mechanisms through *operando* characterization that links activity loss to structural and chemical changes. Materials strategies, such as core-shell structures, gradient electrodes, or protective conductive layers, could enhance stability while maintaining activity. Electrodes should also be tested as complete devices rather than powders alone, with standardized protocols reporting lifetime at fixed current, stability under cycling, and performance recovery after cleaning.

5.5 Practical reactor design

Practical reactor design remains an underdeveloped but essential direction for EGOR||HER coupling. Most current studies are still performed in H-type cells or conventional three-electrode systems, which are useful for catalyst screening and mechanistic analysis but cannot fully represent practical operating conditions. In these static reactors, mass transport is mainly governed by diffusion and stirring, while product accumulation, local pH variation, gas bubble retention, and concentration polarization are difficult to control. As a result, the activity, selectivity, and stability obtained

in H-type cells may not be directly transferable to practical electrolyzers.

For practical applications, EGOR||HER systems need to be evaluated in flow-cell reactors, membrane-electrode assembly systems, or other continuous electrolyzer configurations. Flow-cell reactors can improve EG supply, accelerate product removal, reduce local concentration gradients, and support high-current-density operation. However, both experimental and theoretical studies on EGOR||HER reactor design remain insufficient. Key parameters, such as flow rate, flow-field geometry, electrode spacing, membrane selection, gas-liquid management, pressure drop, and long-term fouling behavior, are rarely optimized systematically. Future work should combine flow-cell experiments with computational fluid dynamics modeling to understand reactant distribution, product transport, and H₂ bubble removal under realistic conditions.

6 Conclusions

In conclusion, EGOR||HER coupled electrolysis provides a promising strategy to integrate PET upcycling with energy-saving hydrogen production. By replacing the sluggish OER with EGOR, this system can lower the overall cell voltage while generating value-added anodic FA/GA products and cathodic H₂. This review summarizes the fundamental EGOR pathways, thermodynamic and energetic descriptors, catalyst design strategies, and reactor configurations for coupled EGOR||HER systems. Catalyst research for EGOR||HER coupling has evolved from precious metals to transition-metal compounds, MOFs, organic catalysts, and hybrid structures, with increasing focus on bifunctionality, selectivity control, and durability. Meanwhile, reactor design is becoming equally important, as mass transport, product crossover, gas-liquid management, and product recovery strongly affect practical performance. Although encouraging progress has been achieved, several challenges remain, including precise C1/C2 selectivity control, efficient downstream separation, catalyst poisoning, real PET-waste impurity tolerance, and the lack of standardized flow-cell evaluation. Future studies should move beyond catalyst-centered H-type cell tests toward integrated device-level optimization, combining *operando* characterization, computational modeling, membrane design, and realistic feedstock evaluation. These efforts will be essential for advancing EGOR-assisted hydrogen production from laboratory demonstrations toward scalable and sustainable PET electro-upcycling technologies.

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Declaration of conflicting interests

The authors declare no conflicting interests regarding the content of this article.

Data availability

All data needed to support the conclusions in the paper are presented in the manuscript and/or the Supplementary Materials.

Additional data related to this paper may be requested from the corresponding author upon request.

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