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Facile Sprayed and Electro-Activated Ultra-Low Pt Loading Catalyst for Hydrogen Evolution Reaction and PEM Water Electrolysis

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ABSTRACT

Polymer electrolyte membrane water electrolysis (PEMWE) is a promising route for high-purity green hydrogen, yet its large-scale deployment is limited by heavy platinum-group-metal (PGM) usage and the high Pt loading required at the cathode. Although methods such as sputtering, atomic layer deposition, and electrodeposition have been explored, they often lead to poor dispersion, non-uniform growth, or agglomeration that reduces catalyst utilization. Achieving high hydrogen evolution reaction (HER) performance at ultra-low Pt loadings, therefore, requires a fabrication strategy capable of producing well-dispersed nanoscale Pt within a thin, mass-transport-efficient catalyst layer. Herein, we introduce a spray-coating assisted electrochemical reduction (Pt-SE) method that forms well-dispersed 2–3 nm Pt nanoparticles at an ultra-low loading of 0.0178 mg_{Pt} cm⁻². Pt-SE delivers excellent HER activity, including an overpotential of 87 mV at 1 A cm⁻² and a mass activity of 34.4 A mg_{Pt}⁻¹, exceeding commercial Pt/C by more than 30-fold. As a PEMWE cathode, Pt-SE achieves 1.62 V and 90.6% higher-heating-value efficiency at 1 A cm⁻². Further, techno-economic analysis shows that the 98% reduction in Pt usage lowers the levelized cost of hydrogen to 3.91\$ kg_{H₂}⁻¹, within the DOE 2030 target range. These results highlight Pt-SE as a scalable and economically compelling approach for ultra-low-PGM PEMWE systems.

1 | Introduction

Hydrogen (H₂) has long been regarded as one of the most sustainable alternatives to fossil fuels for a future low-carbon global economy [1, 2]. However, the majority of hydrogen is still produced from fossil-based processes, which account for more than 2% of global carbon dioxide emissions [3–5]. To achieve decarbonization, a transition toward clean and renewable hydrogen production has become essential. The accelerating global

movement toward carbon neutrality has intensified the demand for clean hydrogen as a sustainable energy carrier [6, 7]. Polymer electrolyte membrane water electrolysis (PEMWE) has emerged as a leading technology for high-purity hydrogen production because of its high current density operation, rapid dynamic response, compact cell architecture, and compatibility with intermittent renewable electricity among diverse water electrolysis systems [8, 9]. Despite these advantages, the widespread deployment of PEMWE remains hindered by its strong dependence

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on platinum-group metals (PGMs), which substantially increase system capital costs [10]. In particular, the cathodic hydrogen evolution reaction (HER) relies almost exclusively on Pt-based catalysts (e.g., Pt on carbon substrate), making Pt utilization one of the most critical cost bottlenecks for large-scale PEMWE systems [11, 12].

Conventional Pt/C catalysts used in acidic HER environments suffer from several intrinsic limitations, including low Pt utilization, particle agglomeration, and the necessity for relatively high Pt loadings (typically 1–2 mg_{Pt} cm⁻² in commercial PEMWE cathodes) [13, 14]. Thick catalyst layers impede mass transport, causing increased resistance to hydrogen diffusion and reduced accessibility of active Pt sites [13, 15]. Various strategies have been investigated to reduce Pt loading while retaining high activity, such as atomic layer deposition, sputtering, and electrodeposition [16–18]. However, these approaches often result in poor dispersion, non-uniform particle size, low scalability, or the formation of large Pt agglomerates, particularly when using electrodeposition methods, where precursor concentration strongly influences particle growth dynamics [19, 20]. Furthermore, on intrinsically rough and porous carbon gas diffusion electrode (GDE), direct electrodeposition from dissolved precursors readily induces non-uniform Pt agglomeration, an issue that becomes particularly detrimental at ultra-low Pt loadings [21, 22]. Moreover, many of these techniques require complex processing steps or specialized equipment, limiting their practicality for large-scale electrolyzer manufacturing. These limitations highlight the need for a robust and scalable synthetic route that can maximize Pt utilization at ultra-low loadings.

Achieving high HER activity with minimal Pt usage requires catalyst architectures that fundamentally differ from those produced by conventional fabrication methods. In particular, uniformly dispersed, ultrasmall Pt nanoparticles are essential for maximizing the electrochemically active surface area while ensuring efficient utilization of every deposited Pt atom. Nanoparticles only a few nanometers in size provide a high density of accessible surface sites, which is crucial for maintaining high catalytic activity even at ultra-low loadings [23–25]. Equally important is the formation of a thin and well-organized catalyst layer, which minimizes mass-transport resistance and facilitates rapid hydrogen diffusion under the high-current-density conditions of typical PEMWE operation [26–28]. Achieving such structural precision requires uniform precursor distribution and a well-regulated electrochemical reduction step, as even small irregularities can trigger particle coalescence and transport limitations that degrade performance [13]. Therefore, a synthesis strategy capable of precisely controlling Pt nucleation and growth, while producing uniform nanostructures at ultra-low loadings, is needed to overcome the limitations of conventional Pt catalyst fabrication on PEMWE cathodes.

Herein, we introduce a spray-coating assisted electrochemical reduction strategy (Pt-SE) designed to overcome the dispersion and agglomeration challenges inherent to conventional Pt deposition. In this method, a pre-distributed Pt precursor layer is first established through spray-coating and subsequently converted into ultrasmall 2–3 nm Pt nanoparticles at an ultra-low loading of 0.0178 mg_{Pt} cm⁻² through electrochemical reduction. To evaluate the impact of this fabrication route, Pt-SE was system-

atically compared with Pt electrodes produced by conventional electrodeposition (Pt-E1 and Pt-E5) in terms of morphology, particle dispersion, and acidic HER activity. These comparisons reveal that well-controlled precursor pre-distribution is essential for suppressing localized nucleation, enabling thin and well-organized catalyst layers that substantially enhance Pt utilization. Benefiting from this architecture, Pt-SE delivers excellent half-cell HER performance, including an overpotential of 87 mV at 1 A cm⁻² and a mass activity of 34.4 A mg_{Pt}⁻¹. Beyond half-cell testing, Pt-SE was independently implemented as a PEMWE cathode, where it achieved 1.62 V at 1 A cm⁻², a mass activity of 82.5 A mg_{Pt}⁻¹ at 1.7 V, and a higher heating value (HHV) efficiency of 90.6% at 1 A cm⁻², notably delivering commercial Pt/C equivalent PEMWE performance while using less than 5% of the Pt loading employed in Pt/C cathodes. A techno-economic analysis further shows that the ultra-low Pt loading reduces the cathode catalyst cost by 98%, lowering the levelized cost of hydrogen to 3.91 \$ kg_{H2}⁻¹, within the DOE-2030 target range.

2 | Results and Discussions

2.1 | Synthesis and Characterization of Catalysts

As illustrated in Figure 1a, Pt-SE was prepared through a two-step synthetic process involving spray coating followed by electrochemical reduction. A Pt precursor solution was sprayed onto the carbon GDE surface placed on a hot plate, forming a pre-distributed precursor layer (Figure S1). After drying under ambient conditions, the precursor-coated GDE was subjected to electrochemical reduction in 0.5 M H₂SO₄ via single linear sweep voltammetry (LSV) scan, converting the Pt precursor into metallic Pt nanoparticles. In contrast, the conventional electrodeposition route used for Pt-E1 and Pt-E5 – electrodeposited Pt catalysts prepared from 1 mM (Pt-E1) and 5 mM (Pt-E5) Pt precursor solutions, respectively—did not include a spray-coating step [17, 29]. Instead, the Pt precursor was directly dissolved in the electrolyte, and the bare GDE was immersed in the solution before applying a single LSV scan to induce Pt deposition, as detailed in the Experimental section. The structural outcomes of these contrasting synthesis strategies were notably different. Pt-E1 and Pt-E5 formed large and non-uniform Pt agglomerates due to localized nucleation during electrodeposition (Figure S2). In contrast, Pt-SE exhibited much smaller and well-dispersed Pt nanoparticles (Figure S2).

To quantitatively assess Pt utilization and material loss during the synthesis, both the initial Pt precursor consumption during the spray-coating step and the dissolved Pt concentration in the electrolyte after electrochemical reduction were considered (Table S1). In the Pt-SE process, approximately 40% of the Pt precursor was lost during spray coating, indicating that not all of the initially supplied precursor was deposited onto the GDE surface. After subsequent electrochemical reduction, only 47.6 µg of Pt was detected in the electrolyte, while 17.8 µg of Pt was finally deposited on the 1 cm² electrode. In sharp contrast, the electrolytes used for Pt-E1 and Pt-E5 contained 7803.4 µg and 39016.8 µg of dissolved Pt, respectively, because the Pt precursor was directly dissolved in the electrolyte before electrodeposition. Nevertheless, the deposited Pt loadings for Pt-E1 and Pt-E5 remained relatively low at 0.0269 and 0.0572 mg_{Pt} cm⁻²,

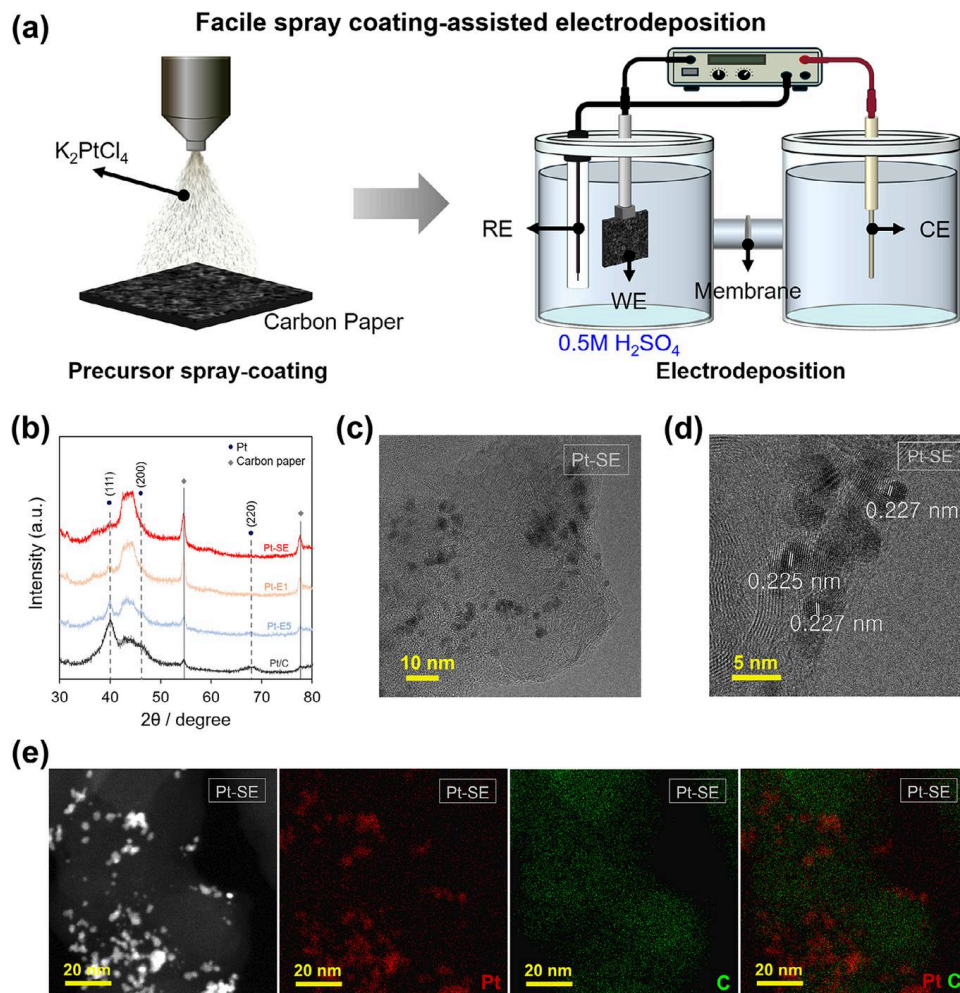


FIGURE 1 | (a) Schematic illustration of the Pt-SE synthesis route, where precursor spray-coating enables precursor pre-distribution on the electrode surface, followed by electrochemical reduction to generate Pt nanoparticles on the electrode surface. (b) XRD patterns of Pt-SE, Pt-E1, Pt-E5, and Pt/C. (c) TEM and (d) HRTEM images of Pt-SE. (e) HAADF-STEM element mappings of Pt-SE.

respectively. Thus, even after considering the precursor loss during the spray-coating step, the amount of unused Pt associated with the Pt-SE process is markedly lower than that in conventional electrodeposition, where a large excess of Pt precursor remains in the bulk electrolyte after deposition. These results demonstrate that, although the spray-coating step still involves precursor loss, the spray-coating-assisted strategy reduces the excessive bulk-electrolyte Pt waste inherent to conventional electrodeposition and provides a more material-efficient route for ultra-low-loading Pt catalyst fabrication.

To further elucidate the structural differences among the catalysts, the XRD patterns of all catalysts were compared with that of commercial Pt/C. For the commercial Pt/C catalysts, a strong Pt (111) peak along with other metallic Pt reflections was clearly observed (Figure 1b). Pt-E1 and Pt-E5 also exhibited the Pt (111) peak, while Pt-E5, which contained a higher Pt loading, showed a noticeably higher peak intensity. However, distinct diffraction peaks were not observed for Pt-SE, which we attribute to its small particle size and extremely low Pt loading ($0.0178 \text{ mg}_{\text{Pt}} \text{ cm}^{-2}$), resulting in insufficient scattering intensity for clear peak formation.

Transmission electron microscopy (TEM) analysis revealed that Pt-SE forms finely dispersed Pt nanoparticles across the GDE surface (Figure 1c). Subsequent quantitative image analysis confirmed a narrow particle size distribution centered at approximately 2–3 nm (Figure S3). Additional TEM images collected from multiple electrode regions consistently showed the similar nanoscale Pt morphology and size distribution, demonstrating that the formation of these ultrasmall Pt nanoparticles is a robust and reproducible outcome of the spray-coating assisted synthesis (Figure S4). For comparison, TEM images of the commercial Pt/C catalyst (Figure S5) reveal similarly sized Pt nanoparticles with comparable nanoscale dispersion to that of Pt-SE, indicating that the Pt domains formed in Pt-SE possess nanoscale dimensions comparable to those of the benchmark Pt/C catalyst.

High-resolution TEM (HRTEM) further revealed well-defined lattice fringes with an interplanar spacing consistent with Pt (111) (Figure 1d). Notably, additional HRTEM analyses of the conventionally electrodeposited Pt-E1 and Pt-E5 catalysts also show similar Pt (111) consistent lattice spacings, indicating that the underlying crystallographic lattice structure is comparable across all catalysts (Figure S6). However, HRTEM lattice fringes

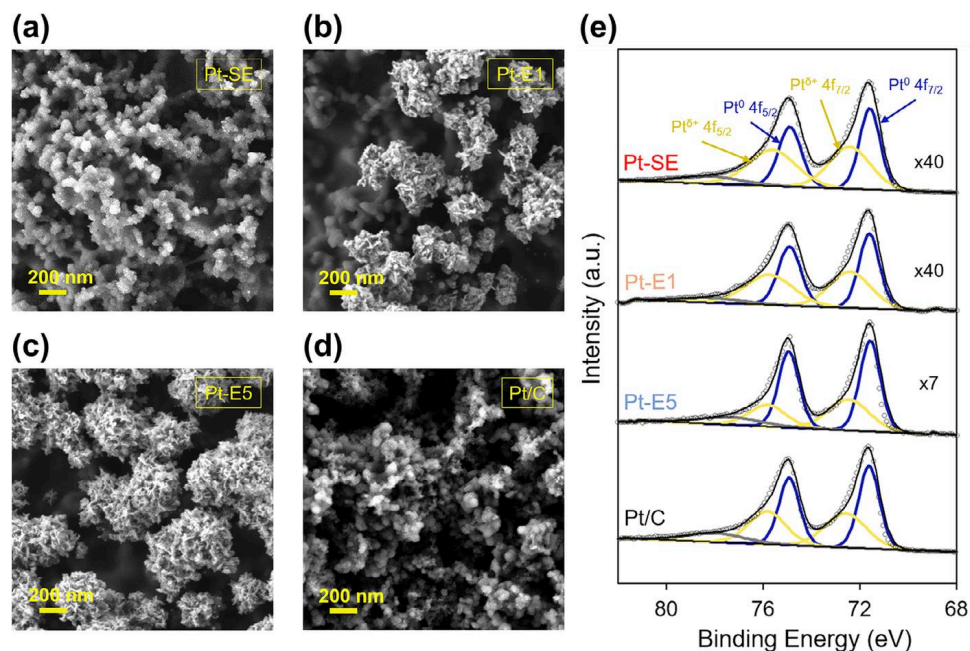


FIGURE 2 | SEM image of (a) Pt-SE, (b) Pt-E1, (c) Pt-E5, and (d) Pt/C. (e) XPS spectra of Pt-SE, Pt-E1, Pt-E5, and Pt/C.

reflect projected crystallographic planes and do not uniquely identify the electrochemically exposed surface facets, particularly for ultrasmall Pt nanoparticles that may expose multiple low-index planes. While the Pt (111) plane is widely regarded as the most thermodynamically stable low-index surface of fcc Pt [30, 31], and few-nanometer-sized Pt nanoparticles are generally known to exhibit high intrinsic HER activity [32], the present results do not imply a uniquely exposed Pt (111) surface. Instead, the key distinction lies in morphology, as Pt-E1 and Pt-E5 exhibit pronounced particle coalescence and agglomeration with much larger Pt domains (Figure S6), whereas Pt-SE retains well-dispersed ultrasmall Pt nanoparticles. Furthermore, high-angle annular dark-field scanning transmission electron microscopy coupled with energy dispersive X-ray spectroscopy (HAADF-STEM EDS) confirmed the post-reduction distribution of these ultrasmall Pt nanoparticles on the carbon GDE surface (Figure 1e), highlighting that the superior catalytic performance of Pt-SE originates from enhanced nanoscale dispersion and maximized electrochemically accessible Pt surface area.

Scanning electron microscopy (SEM) imaging revealed that Pt-E1 and Pt-E5, synthesized by conventional electrodeposition, formed large Pt aggregates on the GDE surface, with particle sizes extending into the hundreds of nanometers (Figure 2b,c). Moreover, the Pt-E5 catalyst, prepared from the higher precursor concentration, exhibited larger agglomerates than Pt-E1. Low-magnification SEM images further confirmed that, while Pt-SE displayed well-dispersed nanoscale Pt features across the GDE, Pt-E1 and Pt-E5 showed highly uneven and irregular Pt deposition (Figure S2). In contrast to these electrodeposited catalysts, Pt-SE consists exclusively of ultrafine Pt nanoparticles dispersed on the carbon GDE, closely resembling the well-dispersed small Pt particles observed in commercial Pt/C (Figure 2a,d). These results highlight that, on rough GDE substrates, direct electrodeposition inherently amplifies localized nucleation and particle

agglomeration, whereas introducing a simple spray-coating step prior to electrochemical reduction effectively suppresses such non-uniform growth by establishing precursor pre-distribution.

The chemical states of the Pt catalysts were analyzed by X-ray photoelectron spectroscopy (XPS), as shown in Figure 2e. All catalysts exhibit characteristic Pt 4f doublets assigned to metallic Pt^0 and partially oxidized $\text{Pt}^{\delta+}$ species originating from the Pt 4f_{7/2} and Pt 4f_{5/2} components. As summarized in Table S2, the Pt^0 4f_{7/2} binding energies of Pt-SE (71.73 eV) and commercial Pt/C (71.74 eV) are slightly higher than those of Pt-E1 (71.61 eV) and Pt-E5 (71.58 eV). Previous studies have consistently reported that Pt nanoparticles in the few-nanometer regime exhibit positive binding-energy shifts relative to bulk Pt, which are primarily attributed to reduced electronic screening and size-dependent final state effects associated with confined metallic domains [33–35]. In contrast, larger Pt particles or agglomerates generally display binding energies close to bulk metallic Pt, reflecting more efficient electronic screening and bulk-like electronic structures [34, 36].

Accordingly, the relatively higher binding energies observed for Pt-SE and Pt/C are consistent with the presence of nanoscale Pt domains, whereas the nearly identical binding energies of Pt-E1 and Pt-E5 reflect their predominantly bulk-like agglomerated structures. The similar binding energies between Pt-SE and Pt/C further indicate that the surface-exposed Pt species probed by XPS experience comparable local electronic environments at the nanoscale, consistent with previous reports showing converged Pt 4f binding energies for carbon-supported Pt catalysts with comparable nanoscale surface domains [35, 37]. Together with the SEM and TEM analyses, these results indicate that the spray-coating assisted process yields finely dispersed nanoscale Pt domains, in contrast to the bulk-like Pt aggregates formed during conventional electrodeposition.

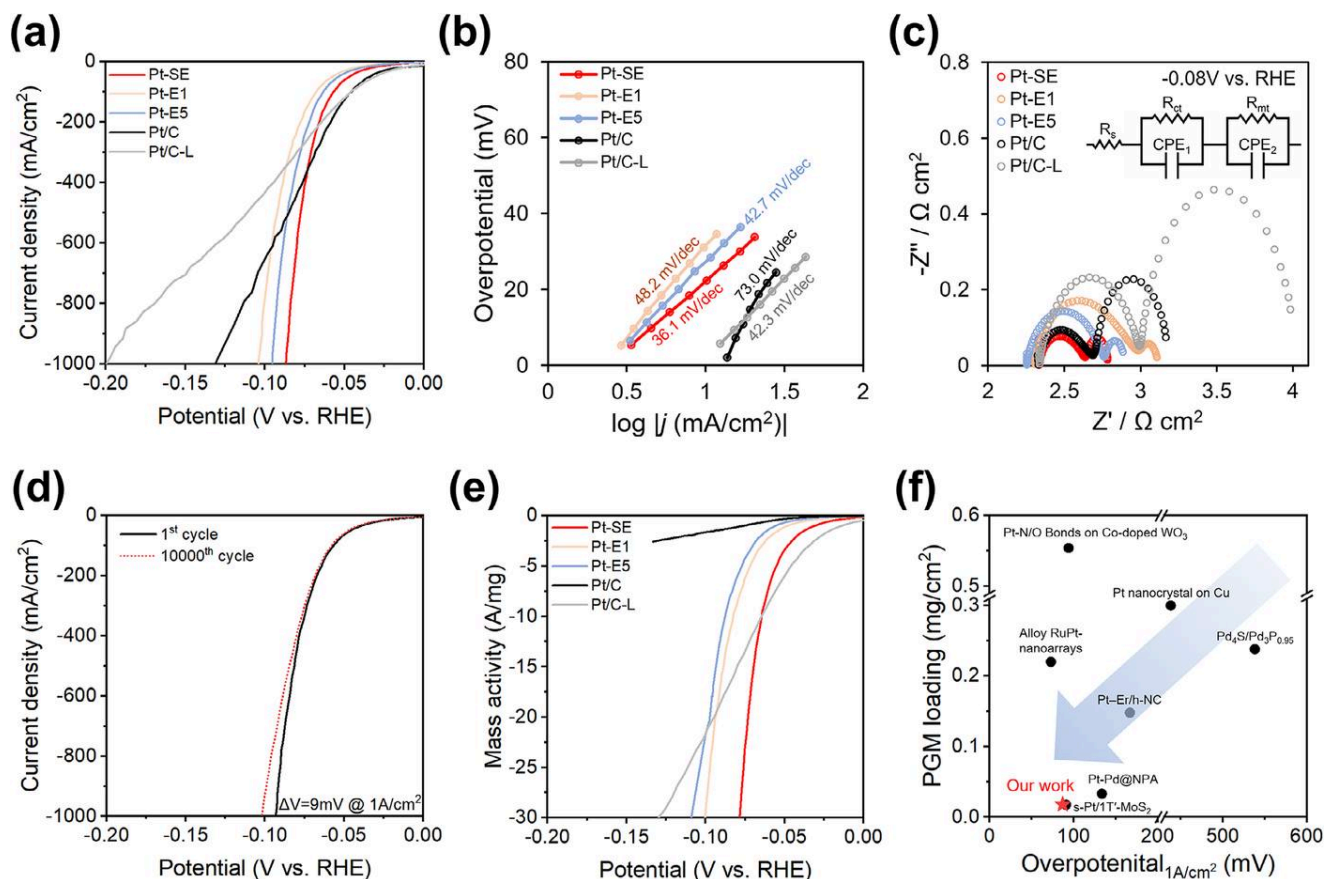


FIGURE 3 | (a) LSV polarization curves and (b) Tafel plots, and (c) EIS comparison for Pt-SE, Pt-E1, Pt-E5, Pt/C, and Pt/C-L. (d) LSV polarization curves for Pt-SE measured before and after 10 000 CV stability test. (e) Pt mass-normalized polarization curves for Pt-SE, Pt-E1, Pt-E5, Pt/C, Pt/C-L. (f) Comparison of the overpotential at 1 A cm⁻² and PGM loading of the Pt-SE with recently reported literature.

2.2 | Electrocatalytic Half-Cell Performance for HER

The HER performance of the synthesized catalysts and commercial Pt/C with different Pt loadings was evaluated in 0.5 M H₂SO₄ at a scan rate of 5 mV s⁻¹. For comparison, a commercial Pt/C electrode with a Pt loading of 0.4 mg_{Pt} cm⁻² was tested, and to enable a more scientifically meaningful comparison with Pt-SE, a low-loading commercial Pt/C electrode, denoted as Pt/C-L, was additionally prepared with a Pt loading of 0.02 mg_{Pt} cm⁻², which is comparable to that of Pt-SE. Figure 3a compares the linear sweep voltammetry (LSV) polarization curves for Pt-SE, Pt-E1, Pt-E5, Pt/C, and Pt/C-L. At a high current density of 1 A cm⁻², Pt-SE exhibited the best HER performance, showing the lowest overpotential of 87 mV, outperforming Pt-E1 (104 mV), Pt-E5 (96 mV), Pt/C (131 mV), and Pt/C-L (200 mV) despite having the lowest Pt loading. The Tafel plots derived from the LSV polarization curves also show that Pt-SE exhibits the lowest value (36.1 mV dec⁻¹), compared with Pt-E1 (48.2 mV dec⁻¹), Pt-E5 (42.7 mV dec⁻¹), Pt/C (73.0 mV dec⁻¹), and Pt/C-L (42.3 mV dec⁻¹) (Figure 3b). The enhanced performance of Pt-SE can be attributed to the formation of well-dispersed nanoscale Pt nanoparticles with a narrow size distribution, which provide a high density of accessible active sites, enabling superior HER activity even at an ultra-low Pt loading. Notably, Pt-SE also outperforms Pt/C-L despite their comparable Pt loadings, indicating that the improved HER activity arises not simply from the amount of

Pt, but from the distinct electrode architecture enabled by the spray-coating-assisted electrodeposition process.

In addition, to evaluate the influence of the electrodeposition protocol employed in the spray-coating assisted synthesis, the HER performances of Pt-SE electrodes prepared under potentiostatic and galvanostatic electrodeposition conditions were compared with those prepared via the LSV-based reduction (Figure S7). The LSV-derived Pt-SE consistently exhibited higher current densities over the entire potential range than those obtained from potentiostatic or galvanostatic electrodeposition, demonstrating that the LSV-based protocol yields superior HER performance under otherwise identical synthesis conditions.

Figure 3c presents the electrochemical impedance spectroscopy (EIS) results measured at -0.08 V vs. RHE for Pt-SE, Pt-E1, Pt-E5, Pt/C, and Pt/C-L. Two semicircles are observed in the Nyquist plots: the semicircle toward the left direction of the x-axis in the high-frequency region corresponds to the charge-transfer resistance (R_{ct}), which represents the kinetic barrier for HER [38, 39], while the semicircle toward the right direction of the x-axis in the low-frequency region is associated with mass-transport resistance (R_{mt}), which reflects limitations in reactant/product transport [40, 41]. Accordingly, the impedance spectra were fitted using a two-time constant equivalent circuit model (Figure 3c), where the high- and low-frequency time constants are commonly associated with interfacial charge-transfer and mass-transport related

impedance responses, respectively [42]. Among the catalysts, Pt-SE exhibited the lowest R_{ct} , followed by Pt/C, Pt-E5, Pt-E1, and Pt/C-L, indicating that Pt-SE achieves the most favorable HER kinetics despite having the lowest Pt loading. The substantially larger impedance response of Pt/C-L further confirms that simply reducing the Pt/C loading to a level comparable to that of Pt-SE does not guarantee comparable catalytic utilization, highlighting the importance of the electrode architecture formed by the spray-coating-assisted electrodeposition process. In contrast, the R_{mt} was similar for Pt-SE, Pt-E1, and Pt-E5, all of which showed significantly lower R_{mt} values than Pt/C. We attribute the higher R_{mt} of Pt/C to its substantially larger Pt loading ($0.4 \text{ mg}_{Pt} \text{ cm}^{-2}$), which produces a much thicker catalyst layer that hinders both the escape of generated hydrogen gas and the supply of water molecules, thereby limiting mass transport [43, 44]. Collectively, these results demonstrate that Pt-SE simultaneously minimizes kinetic and mass-transport resistances, enabling superior HER performance at an ultra-low Pt loading.

To further assess catalyst stability, 10 000 cyclic voltammetry (CV) cycles were conducted between -0.1 and 0.1 V vs RHE. The Pt-SE catalyst exhibited only a 9 mV increase in overpotential at 1 A cm^{-2} after cycling (Figure 3d), indicating excellent electrochemical stability. Post stability test SEM, TEM, and STEM-EDS mapping analyses further confirmed that the Pt-SE maintains well-dispersed ultrasmall Pt nanoparticles (Figure S8), demonstrating the structural robustness of the spray-coating assisted electrodeposition method. In contrast, Pt-E1 and Pt-E5 showed comparatively larger increases of 57 and 20 mV, respectively, under the same conditions (Figure S9), underscoring their inferior stability relative to Pt-SE.

To better compare the catalytic performance, the specific activity of each catalyst was obtained by dividing its geometric current density by the corresponding electrochemically active surface area (ECSA). The ECSA-normalized current density serves as an indicator of the intrinsic activity of a catalyst [45] and was determined using the electric double-layer capacitance (C_{dl}) extracted from the non-Faradaic CV curves (Figure S10). The Pt-SE, Pt-E1, Pt-E5, and Pt/C-L catalysts exhibited substantially lower C_{dl} values of 1.29, 1.36, 1.51, and 1.50 mF cm^{-2} , respectively, compared to 107.5 mF cm^{-2} for Pt/C, which is consistent with their significantly lower Pt loadings (Figure S11). When normalized by ECSA, Pt-SE showed the highest intrinsic activity among all catalysts, followed by Pt-E5, Pt-E1, and Pt/C-L (Figure S12). However, it should be noted that C_{dl} reflects not only the Pt surface area but also the total electrochemically accessible surface area of the entire electrode, particularly for porous carbon-based electrodes. Therefore, to more selectively evaluate the Pt-specific ECSA, hydrogen adsorption/desorption measurements were performed by CV. The hydrogen adsorption/desorption charge (Q_H), which represents the amount of charge associated with reversible hydrogen adsorption on Pt surfaces in the hydrogen underpotential deposition (H_{upd}) region (typically 0–0.4 V vs. RHE), provides a more direct indicator of the electrochemically accessible Pt surface area [46, 47]. As shown in Figure S13, Pt/C displays a much larger H_{upd} peak area and thus a markedly higher Q_H (73.2 mC cm^{-2}) than Pt-SE (3.1 mC cm^{-2}). However, when normalized by the Pt loading, the derived Pt-based ECSA values are comparable, $871.9 \text{ cm}^2 \text{ mg}_{Pt}^{-1}$ for Pt/C and $835.4 \text{ cm}^2 \text{ mg}_{Pt}^{-1}$ for Pt-SE, indicating a similar electrochemically accessible Pt

surface area per unit Pt mass. The substantially larger Q_H of Pt/C therefore primarily originates from its much higher Pt loading, rather than a higher intrinsic dispersion per Pt mass. Notably, despite the similar mass-normalized ECSA, Pt-SE delivers higher HER current densities than Pt/C, suggesting improved apparent intrinsic activity and more effective utilization of Pt sites in the spray-coating-assisted electrodeposition electrode architecture under HER operating conditions.

To highlight the advantage of the ultra-low Pt loading, the performance was also assessed in terms of mass activity, which normalizes current to Pt mass loading. Pt-SE exhibited significantly higher mass activity than the other catalysts, showing a particularly large difference compared with commercial Pt/C (Figure 3e). At -0.08 V vs. RHE, Pt-SE delivered a mass activity of 34.4 A mg^{-1} , which is more than 30 times higher than that of Pt/C (1.1 A mg^{-1}) and approximately two, four, and five times that of Pt/C-L (15 A mg^{-1}), Pt-E1 (8.7 A mg^{-1}), and Pt-E5 (6 A mg^{-1}), respectively (Figure S14). In the 2D plot of HER overpotential at 1 A cm^{-2} versus platinum-group-metal (PGM) loading, our Pt-SE catalyst is positioned in the lower-left region, demonstrating its superior performance relative to previously reported PGM catalysts (Figure 3f). Detailed information on other recently reported literature is provided in Table S3.

These results reveal an electrode-architecture-driven structure-activity relationship in which spray-coating-assisted precursor pre-distribution suppresses localized Pt nucleation on the rough GDE, thereby forming well-dispersed nanoscale Pt domains with improved electrolyte accessibility and electrical connection to the conductive substrate. This architecture lowers charge-transfer resistance and mitigates catalyst-layer-induced transport limitations, thereby enhancing apparent Pt utilization at an ultra-low Pt loading [26, 28]. Consequently, Pt-SE exhibits outstanding HER performance, including the lowest overpotential, smallest Tafel slope, superior mass-transport characteristics, and remarkably high mass activity among all examined catalysts. The comparison with Pt/C-L further confirms that the superior activity of Pt-SE arises from enhanced Pt utilization enabled by its electrode architecture, rather than merely from reduced Pt loading, underscoring Pt-SE as a highly efficient and PGM-economical catalyst for acidic HER.

2.3 | Electrocatalytic Full Cell Performance for PEMWE

The Pt-SE catalyst was further validated as a cathode in the PEMWE system, paired with an IrO_x -coated Ti felt anode (1 mg cm^{-2}) under operating conditions of 80°C using DI water as the electrolyte (Figure 4a). For comparison, a Pt/C cathode ($0.4 \text{ mg}_{Pt} \text{ cm}^{-2}$) was also tested with the same anode configuration. Hereafter, PEMWE systems are denoted as cathode // anode. The polarization curves show that the Pt-SE cathode delivers performance comparable to that of Pt/C. Specifically, the Pt-SE // IrO_x -Ti felt required voltages of 1.55 and 1.62 V to reach current densities of 0.5 and 1 A cm^{-2} , respectively, while the Pt/C // IrO_x -Ti felt required 1.54 and 1.62 V to achieve the same current densities (Figure 4b). For durability evaluation, the Pt-SE // IrO_x -Ti felt was tested at a constant current density of 1 A cm^{-2} and 50°C , a temperature within the typical PEMWE operating

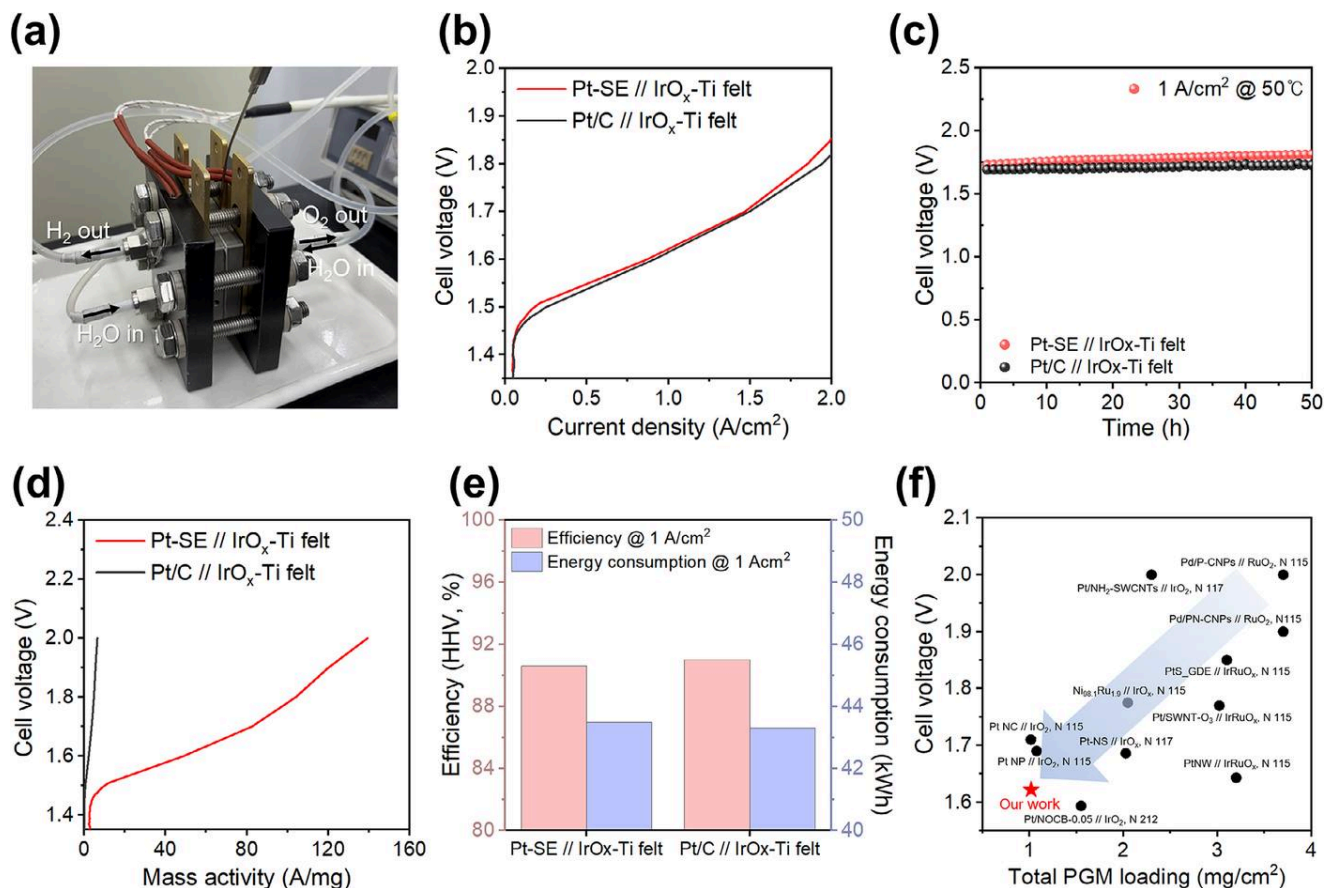


FIGURE 4 | (a) The photograph of the PEMWE full cell device. (b) PEMWE polarization curves for Pt-SE // IrO_x-Ti felt, and Pt/C // IrO_x-Ti felt. (c) Stability test for Pt-SE // IrO_x-Ti felt, and Pt/C // IrO_x-Ti felt at 1 A cm⁻² and 50°C. (d) Pt mass-normalized polarization curves for Pt-SE // IrO_x-Ti felt, and Pt/C // IrO_x-Ti felt. (e) Energy efficiency (higher heating value, HHV) and the corresponding energy consumption at 1 A cm⁻² for Pt-SE // IrO_x-Ti felt, and Pt/C // IrO_x-Ti felt. (f) Comparison of total PGM loading and cell voltage at 1 A cm⁻² with recently reported cathode-modified literature.

range, chosen to minimize temperature-accelerated membrane degradation and thereby better isolate the stability of the ultra-low-Pt cathode architecture [48, 49]. Under these conditions, the Pt-SE // IrO_x-Ti felt exhibited a cell-voltage decay rate of 1.66 mV h⁻¹ over 50 h of operation (Figure 4c). Although this degradation rate is slightly higher than that of the commercial Pt/C cathode measured under identical conditions, this behavior can be attributed to the durability trade-off associated with the ultra-low Pt cathode architecture. Because the Pt loading of Pt-SE is only 0.0178 mg_{Pt} cm⁻², each active Pt domain is expected to sustain a higher local current burden at the same geometric current density. Moreover, the sparse Pt nanoparticle network may be more sensitive to loss of lateral electrical connectivity, while partial particle migration or local agglomeration can induce a larger relative voltage increase than in a high-loading Pt/C catalyst layer; similar degradation behavior has been reported for ultra-low Pt PEMWE cathodes [50]. At the same time, post-durability SEM and HAADF-STEM EDS mapping show that Pt species remain broadly distributed on the carbon GDE after PEMWE operation, suggesting that the Pt-SE cathode retains its electrode-scale structural integrity despite these possible local-scale degradation pathways (Figure S15). Further durability improvement may be achieved by optimizing the cathode catalyst-layer architecture, such as introducing an appropriate ionomer content to improve proton-conducting pathways and

catalyst binding [26, 51], and by strengthening Pt anchoring to suppress particle migration/agglomeration [24].

To further assess its catalytic advantages, the PEMWE mass activity was evaluated based on Pt loading. Pt-SE // IrO_x-Ti felt exhibited mass activities of 48.9 and 82.5 A mg⁻¹ at 1.6 and 1.7 V, respectively, whereas Pt/C // IrO_x-Ti felt showed values of only 2.3 and 3.7 A mg⁻¹ under the same conditions (Figure 4d). Accordingly, Pt-SE achieves more than a 20-fold enhancement in mass activity compared with Pt/C, demonstrating that comparable PEMWE performance can be achieved with only 4.5% of the Pt loading used in Pt/C. These results highlight the compensatory advantages of Pt-SE in other performance dimensions, including markedly enhanced Pt utilization, substantially reduced cathodic Pt usage, and Pt/C comparable initial PEMWE efficiency. Figure 4e summarizes energy efficiency (higher heating value, HHV) and the corresponding energy consumption at 1 A cm⁻². Energy efficiency is defined as the ratio between the thermodynamic energy content of hydrogen (39.4 kWh kg_{H₂}⁻¹ on an HHV basis) and the actual electrical energy input. Accordingly, energy consumption represents the amount of electricity required to produce 1 kg of H₂ at the measured cell voltage and is calculated as 39.4 kWh kg_{H₂}⁻¹ divided by the cell efficiency. Based on this definition, the Pt-SE // IrO_x-Ti felt cell achieves an efficiency of 90.6% at 1 A cm⁻², a practically relevant operating current

density for a real PEMWE system [52–54], corresponding to an energy consumption of 43.5 kWh kg_{H2}⁻¹. This performance significantly outperforms state-of-the-art commercial water electrolyzers, which typically require ≈53 kWh kg_{H2}⁻¹ [55], and is already approaching the IRENA 2050 target of <42 kWh kg_{H2}⁻¹ [55]. Notably, this near-target energy consumption is achieved with a total PGM loading of only 1.0178 mg cm⁻², representing a substantial reduction relative to conventional PEMWE systems.

Finally, our Pt-SE // IrO_x-Ti felt system was benchmarked against previously reported PEMWE devices using a 2D map of total PGM loading (including both cathode and anode) versus cell voltage at 1 A cm⁻². With a total PGM loading of 1.0178 mg cm⁻² and a low operating voltage of 1.62 V, it demonstrates a competitive combination of low total PGM loading and high cell efficiency among previously reported cathode-modified PEMWE systems (Figure 4f). Detailed information on other recently reported literature is provided in Table S4. Overall, Pt-SE demonstrates excellent PEMWE performance despite its ultra-low Pt loading, delivering competitive cell voltages, outstanding mass activity, and high energy efficiency. While further durability optimization is required before long-term practical deployment, the present results demonstrate that Pt-SE is a highly Pt-efficient cathode platform with a favorable cost-performance balance, rather than a fully durability-optimized commercial cathode. Its superior catalyst utilization enables substantial PGM reduction while maintaining Pt/C-comparable initial PEMWE performance, positioning Pt-SE as an economically promising ultra-low-Pt cathode architecture for future PEMWE development.

2.4 | Techno-Economic Analysis of PEMWE System

A techno-economic analysis (TEA) was conducted to assess the economic feasibility of the proposed Pt-SE cathode-based commercial-scale PEMWE system by calculating the LCOH. We assumed a 1 MW-scale facility based on DOE and Energy Information Administration (EIA) reports. The system's capital expenditure (CAPEX) was set at 1850 \$ kW⁻¹ [56], reflecting current commercial electrolyzer costs, and the electricity price was assumed to be 0.03 \$ kWh⁻¹ [57], assuming electricity sourced from renewable energy. Detailed information about TEA is available in Supporting Information. We investigated the effect of current density on LCOH and identified the corresponding optimal operating point for the PEMWE system (Figure 5a). As the current density increases, the power output per cell rises, reducing the number of cells and stacks required to achieve the 1 MW design capacity, and thereby lowering the CAPEX.

In contrast, operating expenditure (OPEX) shows a two-stage behavior driven by the competing effects of electrolyzer size and electricity consumption. At low operating current densities below 1 A cm⁻², increasing the current density decreases the total cell area required to meet the fixed 1 MW production capacity. This reduces the number of cells and stacks, which lowers the balance-of-plant (BOP) operation requirements and consequently drives a gradual decrease in OPEX. However, at operating current densities above 1 A cm⁻², the rise in cell voltage reduces the cell efficiency, meaning that more electrical energy is required for producing hydrogen. Because electricity accounts for the majority

of OPEX (Figure S16), this efficiency loss outweighs the benefits of reduced cell area and results in an overall increase in OPEX at higher current densities.

The interaction between these factors produces a convex LCOH profile with a global minimum at 1 A cm⁻². At this point, the LCOH reaches 3.91 \$ kg_{H2}⁻¹, which is more than \$1 lower than the DOE's 2024 LCOH range for commercially deployed PEMWE systems (5.2–5.7 \$ kg_{H2}⁻¹) (Figure 5a) [56]. Moreover, this value lies within the DOE's projected 2030 target range (3.1–4.2 \$ kg_{H2}⁻¹) [56]. One factor contributing to the reduced LCOH is the substantial decrease in the cathode cost. As a result of the low catalyst loading adopted in our study, the catalyst cost per single cell was dramatically reduced from 0.04 \$ cm⁻², based on the National Renewable Energy Laboratory (NREL) report [58], to 0.0007 \$ cm⁻², corresponding to a 98% reduction (Figure 5b). This intrinsic improvement highlights the important role of material and electrode design innovations in lowering hydrogen production cost, while also emphasizing that broader system-level cost reductions remain essential.

Beyond this baseline analysis, we conducted a sensitivity analysis to examine how two dominant external economic factors, system CAPEX per kW and electricity price in OPEX, affect overall hydrogen costs. When applying the DOE-projected CAPEX values for 2030 (1500 \$ kW⁻¹) and 2050 (1125 \$ kW⁻¹) [59], the resulting LCOH decreases accordingly (Figure 5c). Under the optimistic ultimate target of 150 \$ kW⁻¹, the LCOH falls below 3 \$ kg_{H2}⁻¹, clearly indicating that system CAPEX is the dominant contributor to the reduction of LCOH (Figure 5c). As electricity prices decrease, the LCOH correspondingly declines, and the optimal operating point shifts toward higher current densities because the economic disadvantage associated with increased voltage and power consumption is mitigated. Under the extreme scenario of 0 \$ kWh⁻¹, representing surplus or curtailed renewable electricity, the LCOH approaches the DOE's aspirational 1 \$ kg_{H2}⁻¹ target [59] even when the CAPEX remains at the current commercial level of 1850 \$ kW⁻¹ (Figure 5d). At this point, the OPEX contribution becomes negligible, and system CAPEX emerges as the primary cost driver (Figure 5d). These results collectively demonstrate that achieving truly low-cost green hydrogen requires simultaneous reductions in both system CAPEX and electricity price in OPEX.

3 | Conclusion

In summary, we developed a spray-coating assisted electrochemical reduction strategy that enables the formation of well-dispersed 2–3 nm Pt nanoparticles at an ultra-low loading of 0.0178 mg_{Pt} cm⁻². Systematic comparison with conventionally electrodeposited Pt electrodes revealed that precursor pre-distribution is essential for suppressing localized nucleation and agglomeration, thereby producing a thin, well-organized catalyst layer with superior accessibility of active sites. Accordingly, by introducing a simple spray-coating-assisted precursor pre-distribution step, the present approach effectively mitigates non-uniform Pt agglomeration during direct electrodeposition on rough GDE substrates and enables improved Pt utilization at ultra-low loadings. Benefiting from this architecture, Pt-SE

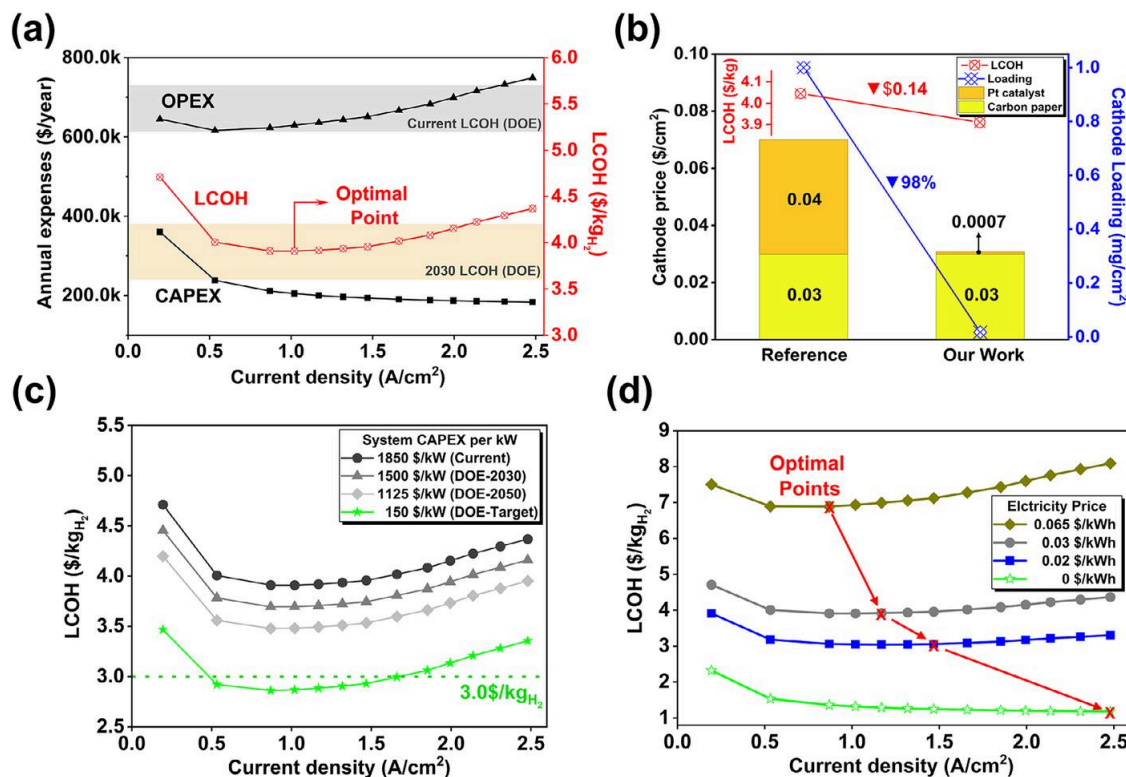


FIGURE 5 | (a) Variation of CAPEX, OPEX, and LCOH with current density, indicating the resulting optimal operating point. (b) Impact of single cell cathode price and loading on the reduction of LCOH. (c) Sensitivity analysis of the Levelized Cost of Hydrogen (LCOH) to variations in system CAPEX and (d) electricity prices; “DOE-2030” and “DOE-2050” denote the projected CAPEX values reported by DOE.

delivers outstanding acidic HER performance, including an overpotential of only 87 mV at 1 A cm⁻², along with the lowest Tafel slope and charge-transfer resistance among all tested catalysts, as well as excellent operational stability. When applied as a PEMWE cathode, the Pt-SE // IrO_x-Ti felt system delivers a cell voltage of 1.62 V and an HHV efficiency of 90.6% at 1 A cm⁻², along with a mass activity of 82.5 A mg⁻¹ at 1.7 V. These results demonstrate that high-efficiency PEMWE operation can be achieved with less than 5% of the Pt loading used in commercial Pt/C cathodes, underscoring the remarkable Pt utilization achieved by the newly developed spray-coating assisted method. Techno-economic analysis further confirms the practical advantages of our cathode fabrication approach: the ultra-low Pt loading reduces the single-cell cathode catalyst cost by 98% and lowers the LCOH to 3.91 \$ kg_{H₂}⁻¹, comfortably within the DOE’s projected 2030 target range. Overall, our results provide an experimentally validated and economically advantageous breakthrough toward the development of an ultra-low PGM-based efficient PEMWE system.

4 | Experimental Section

4.1 | Chemicals

All chemicals were used directly without any further purification. Potassium tetrachloroplatinate (II) (K₂PtCl₄, 99.9%), Pt/C (40 wt%), Nafion 117 solution (5wt%, EW 1000), sulfuric acid (H₂SO₄, 95–98%) were purchased from Sigma Aldrich. Ethanol (99.95%) was purchased from Thermo Fisher Scientific. Ar gas (99.999%)

was purchased from Seoul Speciality Gases Co., Ltd. Deionized water (DI water, 18.2 MΩ cm⁻¹) was used throughout all the experiments.

4.2 | Synthesis of Pt-SE, Pt-E1, and Pt-E5 Electrodes

Pt-SE was synthesized through a spray coating method followed by electrochemical reduction. Specifically, K₂PtCl₄ (0.0415 g) was dissolved in DI water (9.5 mL) and ethanol (9.5 mL), and the precursor solution was sonicated to ensure complete dissolution. It was observed that when the precursor concentration exceeded this optimized value, premature reduction of the Pt precursor occurred in the solution, leading to the formation of black particulate aggregates; therefore, the concentration was carefully controlled to avoid unintended particle nucleation. The solution was spray-coated at a flow rate of 1 mL min⁻¹ onto a gas diffusion electrode (GDE, Sigracet 39BB) placed on a hot plate maintained at 100°C. These spray-coating conditions were selected to promote rapid evaporation of the precursor solution after spraying and to suppress local precursor accumulation during drying. The GDE was used as received without any surface pretreatment. After drying under ambient conditions, the precursor-coated GDE was immersed in 40 mL of 0.5 M H₂SO₄ and subjected to electrochemical reduction in H-cell using a single linear sweep voltammetry (LSV) scan from 0.2 V vs RHE to the potential corresponding to a current density of 1000 mA cm⁻² at a scan rate of 5 mV s⁻¹ to convert Pt precursors into metallic Pt. To evaluate the influence of the electrodeposition

mode in the spray-coating-assisted process, additional reductions were performed using potentiostatic and galvanostatic protocols. For potentiostatic electrodeposition, chronoamperometry (CA) was conducted at constant potentials of -0.1, -0.2, and -0.3 V vs RHE for 60 s. For galvanostatic electrodeposition, chronopotentiometry (CP) was conducted at constant current densities of -100, -500, and -1000 mA cm⁻² for 60 s. The reduction duration of 60 s was selected to be comparable to the effective reduction time required for the LSV-based process, thereby enabling a fair comparison among different reduction protocols. Pt-E1 was synthesized via a conventional electrodeposition method. Bare GDE was immersed in 40 mL of 0.5 M H₂SO₄ with 1 mM K₂PtCl₄, followed by electrodeposition in H-cell through a single LSV scan from 0.2 V vs. RHE to the potential corresponding to a current density of 1000 mA cm⁻² at a scan rate of 5 mV s⁻¹. Pt-E5 was synthesized in the same way as Pt-E1, except that 5 mM K₂PtCl₄ was used instead of 1 mM. The Pt loading mass of the electrode is 0.0178, 0.0267, and 0.0572 mg cm⁻² for Pt-SE, Pt-E1, and Pt-E5, respectively.

4.3 | Preparation of Control Electrodes Based on the Commercial Pt/C

200 mg of commercial 40 wt% Pt/C was homogeneously dispersed into a mixed solution of DI water (9.8 mL), isopropanol (9.8 mL), and Nafion ionomer (400 µL) by ultrasonic processing for 1 h. The resulting catalyst ink was spray-coated onto the gas diffusion electrode placed on a 100°C hot plate using a spray coater and dried in air, with the Pt loading controlled at 0.4 mg cm⁻². To enable a fair comparison with the Pt-SE electrode, a low-loading Pt/C control electrode, denoted as Pt/C-L, was additionally prepared using the same commercial Pt/C ink. The Pt loading of Pt/C-L was precisely controlled to 0.02 mg cm⁻² by adjusting the sprayed ink volume with the spray coater.

4.4 | Characterization of Electrode Materials

Grazing incidence X-ray diffraction (GIXRD) measurements were performed on Smartlab (Rigaku) with Cu-Kα radiation ($\lambda = 1.5418$ Å) using a grazing incidence mode, at an angle of incidence of 0.5 degrees to ensure surface sensitivity of electrode. Scanning electron microscopy (SEM, GeminiSEM 560) was used to examine the electrode surfaces of the samples. Transmission electron microscopy (TEM) imaging was performed on a field emission high resolution microscope (JEM-F200) operating at 200 kV. For TEM sample preparation, the Pt-deposited surface of the GDE was mechanically scraped, and the collected material was dispersed in ethanol. The suspension was subsequently ultrasonicated to achieve uniform dispersion of the detached particles, followed by drop-casting onto a TEM grid and drying under ambient conditions before TEM observation. Energy-dispersive X-ray spectroscopy (EDS) element mapping was carried out on JEM-F200 at 200 kV using Au TEM grids. X-ray photoelectron spectroscopy (XPS) measurements were performed on AXIS-HSi spectrometer with Al Kα ($h\nu = 1486.6$ eV) as the exciting source. The energies of each element were calibrated by adventitious C1s (284.8 eV). An inductively coupled plasma mass spectrometer (ICP-MS, PlasmaQuant MS Elite) was used to quantify the Pt loading mass on each electrode.

4.5 | Electrochemical Measurements

The electrochemical measurements were performed in a three-electrode configuration controlled by an electrochemical workstation (Gamry Interface 1000, Gamry Instruments). An Ag/AgCl electrode (3 M NaCl) and Pt wire were used as the reference and counter electrodes, respectively. Electrochemical performance tests were conducted in a one-compartment cell using 0.5 M H₂SO₄ as the electrolyte. The electrolyte was purged with Ar gas for at least 20 min to remove residual air. All applied potentials were converted to the reversible hydrogen electrode (RHE) potentials by the relation $E_{\text{RHE}} = E_{\text{Ag/AgCl}} + 0.197 + 0.05916 \times \text{pH}$ with IR compensation. The LSV polarization curves were recorded from 0 to -0.5 V vs. RHE with a scan rate of 5 mV s⁻¹. The electrochemical impedance spectroscopy (EIS) plots were recorded over a frequency range of 100 kHz to 10 mHz with an AC amplitude of 10 mV. The stability tests were conducted by applying the cyclic voltammetry (CV) sweeps between 0.1 and -0.1 V (vs RHE) at a scan rate of 100 mV s⁻¹ for 10 000 cyclic voltammetry cycles in an Ar-saturated 0.5 M H₂SO₄ electrolyte. The electrochemically active surface area (ECSA) was determined by performing CV in the non-Faradaic potential region (0.10–0.20 V vs RHE) at scan rates of 20, 40, 60, 80, and 100 mV s⁻¹. The double-layer capacitance (C_{dl}) was obtained by plotting $\Delta j/2$ as a function of the scan rate, where $\Delta j/2$ represents the difference in current density between the anodic and cathodic sweeps. The ECSA was then calculated using $\text{ECSA} = C_{\text{dl}} / C_s$, where C_s is the specific double-layer capacitance, assumed to be 0.04 mF cm⁻². Hydrogen adsorption/desorption measurements were conducted by CV over a potential range of 0–1.4 V vs. RHE at a scan rate of 5 mV s⁻¹ in 0.5 M H₂SO₄. The active area of the electrodes was 1 cm².

4.6 | Performance Evaluation of the PEMWE

All cell tests were performed in a PEM electrolyzer cell with an active area of 1 cm². The operating temperature was maintained at 80°C unless otherwise specified. DI water was supplied to both the cathode and anode at a flow rate of 10 mL min⁻¹, and the pressure on both sides was kept at 1 atm. The prepared Pt-SE or Pt/C electrode was used as the cathode, while 1 mg cm⁻² IrO_x-coated Ti felt served as the anode. Nafion 212 was employed as the membrane. All performance measurements were controlled using an electrochemical workstation (ZIVE SP1 with SP1+ Booster), and no IR compensation was applied.

Author contributions

Dongwook Oh: writing – review and editing, resources, investigation. **Donggeun Eom:** conceptualization, methodology, writing – review and editing, writing – original draft. **Hyoungsoon Lee:** writing – review and editing, funding acquisition, visualization. **Youngeob Lee:** conceptualization, methodology, writing – review and editing. **Sangwook Park:** conceptualization, writing – review and editing, funding acquisition, visualization, project administration.

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Conflicts of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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Supporting Information

Additional supporting information can be found online in the Supporting Information section.

Supporting File: sml74847-sup-0001-SuppMat.docx.