

PEM Water Electrolysis: Cell-Level Considerations for Gigawatt-Scale Deployment

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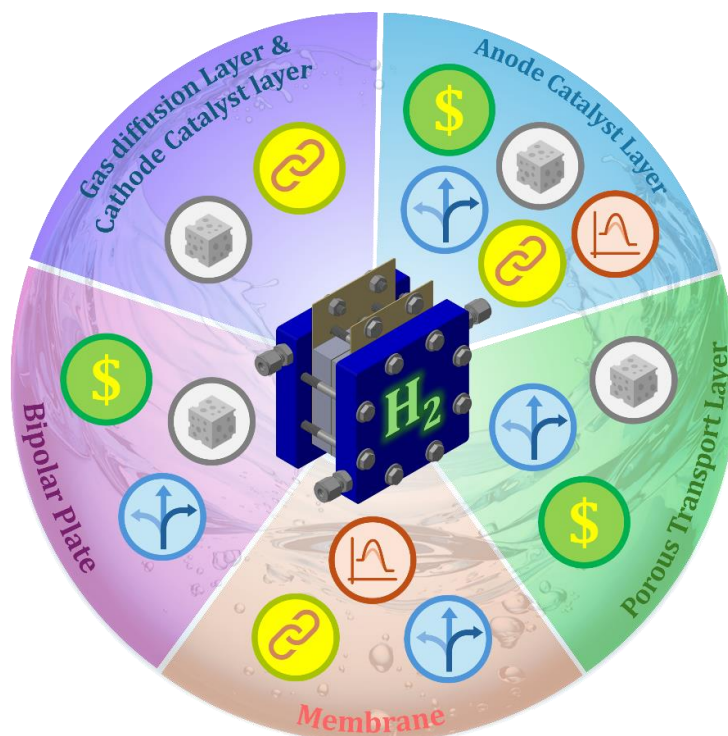
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Abstract

Hydrogen produced with no greenhouse gas emissions is termed “green hydrogen” and will be essential to reaching decarbonization targets set forth by nearly every country as per the Paris Agreement. Proton exchange membrane water electrolyzers (PEMWEs) are expected to contribute substantially to the green hydrogen market. However, PEMWE market penetration is insignificant, accounting for less than a gigawatt of global capacity. Achieving substantive decarbonization via green hydrogen will require PEMWEs to reach capacities of hundreds of gigawatts by 2030. This paper serves as an overarching roadmap for cell-level improvements necessary for gigawatt-scale PEMWE deployment, with insights from three well-established hydrogen technology companies included. Considerations for economies of scale, renewable energy prices, governmental policies, durability testing, and component-specific improvements will be presented, with focus centralized on necessary scientific research goals.



Keywords: Green hydrogen; Water electrolysis; PEM water electrolyzers; Membrane electrode assembly; Electrocatalysts; Iridium; Durability; Degradation

1. Introduction

Nearly every country has joined the landmark Paris Agreement to achieve global net zero emissions by 2050. Hydrogen will be essential to reach these targets as it can mitigate emissions for sectors without promising decarbonization solutions, such as grid-scale energy storage, long-haul transportation, and industrial manufacturing.^{1,2} It can be used electrochemically to generate electricity or as a chemical feedstock to produce commodities such as steel, cement, ammonia, and other valued chemicals.³ Since hydrogen usage does not generate greenhouse gases (GHG), as long as the energy used to generate hydrogen comes from clean power sources (e.g., solar, wind, geothermal, and nuclear), the hydrogen can be deemed “green.”

A proton exchange membrane water electrolyzer (PEMWE) is a promising green hydrogen technology that utilizes electricity to split water into oxygen and hydrogen. When paired with clean power sources, PEMWEs can mass-produce green hydrogen and are expected to make up 40% of the green-hydrogen market, with the remaining 60% composed of alkaline and solid oxide electrolysis.⁴ However, market penetration of green hydrogen is insignificant compared to GHG-intensive hydrogen generation methods due to high production costs (\$4.0-\$9.0/kg vs. \$1.0-\$2.5/kg).⁵ As of 2022, global green hydrogen capacity was 11 GW (1GW can produce ~500 tons H₂ daily⁶),⁷ with PEMWEs making up only 0.3 GW. To achieve net zero emissions by 2050, it is projected that PEMWE capacity will need to reach 220 GW by 2030 and 1130 GW by 2050.⁴

1.1 PEMWE Overview

Three types of water electrolyzers are expected to make up the green hydrogen market: alkaline, solid oxide, and proton exchange membrane. While alkaline electrolysis—characterized by having two electrodes operating in a liquid alkaline electrolyte—has become fully commercialized, its inability to operate under highly dynamic loads and at high current densities and pressures restricts its potential for large-scale production.^{8,9} Solid oxide electrolyzers—characterized by high operating temperatures (>500°C) and the use of a ceramic electrolyte—are highly efficient and may hold promise. However, its developmental stage is still early,⁹ with only a handful of companies selling these products. We refer the reader to the literature for detailed analyses of the different electrolyzer types.^{8–12} PEMWEs possess several characteristics that make them suitable for widespread green hydrogen production: high efficiencies, long lifetimes, large range of operational current densities (up to 3 A cm⁻²)^{13,14}, fast dynamic response times, and ability to operate at differential pressure.⁹

A generic schematic of a PEMWE cell is shown in **Figure 1**. Liquid water is fed into the anode side where the oxygen evolution reaction (OER) occurs, generating oxygen gas bubbles, protons, and electrons. The oxygen exits the cell while the electrons and hydrogen ions (which carry water via electro-osmotic drag)¹⁵ travel—via current collectors and the membrane, respectively—to combine at the cathode, where the hydrogen evolution reaction (HER) occurs to generate hydrogen gas.

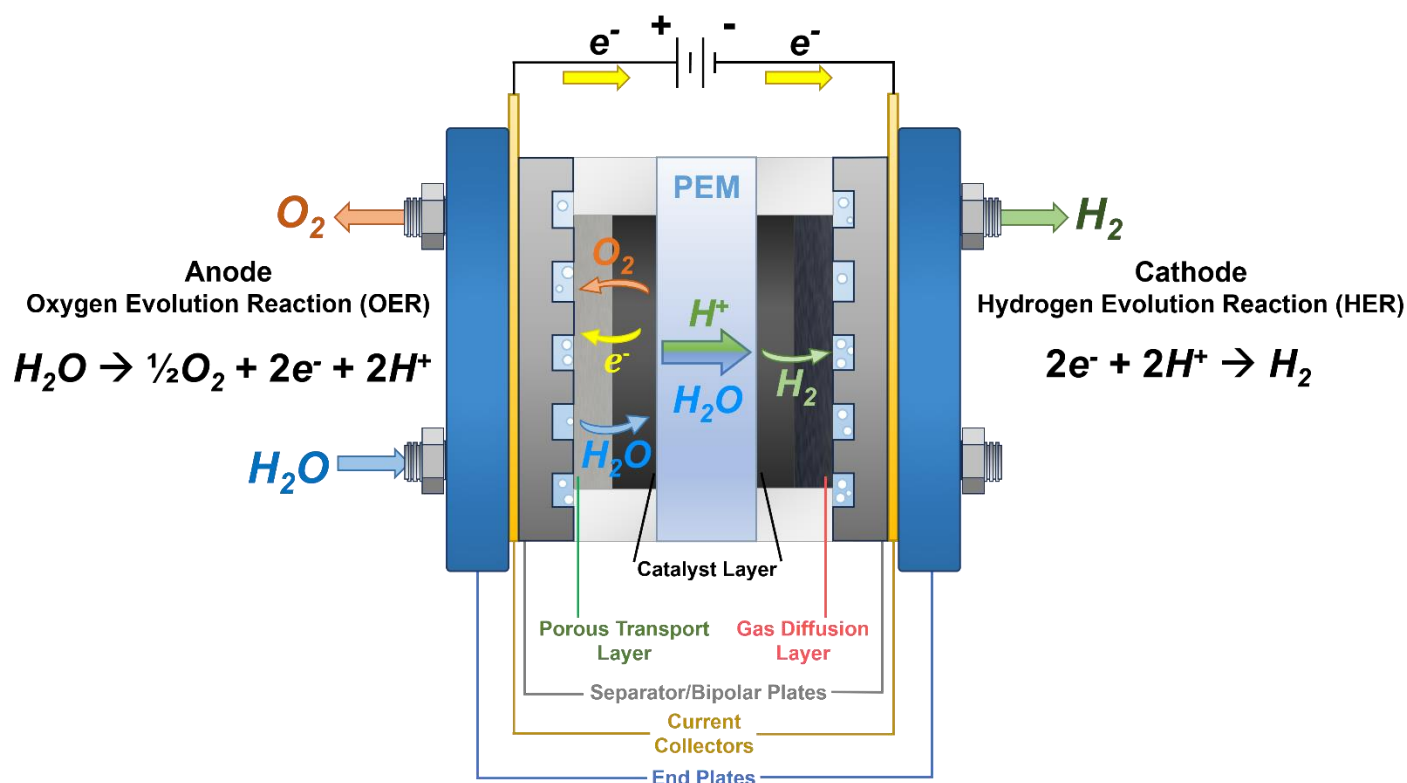


Figure 1. Generic schematic of a proton exchange membrane water electrolyzer (PEMWE).

A PEMWE cell consists of a proton exchange membrane (PEM), two catalyst layers, two transport layers, and two separator plates. The PEM ($\sim 100 \mu m$)¹⁶ is responsible for maximizing protonic conductivity while compartmentalizing the oxygen gas to the anode and hydrogen gas to the cathode. Since gas permeability is inversely correlated with ionic conductivity,¹⁷ achieving low permeability while keeping conductivity high is a hallmark challenge. Sandwiching the membrane are two catalyst layers ($\sim 1-10 \mu m$)¹⁸ which serve as the electrodes of the system—typically consisting of an iridium-based catalyst for the anode and a platinum-based catalyst for the cathode.⁸ Outside each catalyst layer are diffusion layers: the gas diffusion layer (GDL, $100-300 \mu m$)¹⁹ on the cathode side and the porous transport layer (PTL, $\sim 250-500 \mu m$)²⁰ on the anode side. The GDL is primarily responsible for gaseous transport (hydrogen gas out), while the PTL is responsible for liquid and gaseous transport (liquid water in; oxygen gas out). Both the PTL and GDL require establishing electronic conduction with the adjacent catalyst layer, but the PTL is subject to much higher potentials, necessitating material limitations.⁸ The last component on each side of the cell is the separator plate ($\sim 1.5 mm$)²¹ which contains flow pathways to distribute reactants (water) and remove products (oxygen and hydrogen gas).²² Please note that in a stack, numerous cells are combined in series, so separator plates will contain flow fields on both sides (unless they are located at the ends). Thus, due to their two-sided nature, separator plates are also called bipolar plates (BPs). Commercial stacks contain many cells ($\sim 30-200$)^{23,24} each with active areas of $\sim 1000-3000 cm^2$, with total power capacity between $\sim 500 kW-3 MW$.

Although hydrogen is the primary product of PEMWEs, oxygen can also have valuable uses.²⁵ Interestingly, some PEMWE designs even specialize in oxygen production.²⁶ The oxygen can be used in medicine (heart/lung disease patients), defense (fighter aircraft and submarines), spacecraft (life support²⁷), and industry (food processing, welding, metallurgy, semiconductors), to name a few. In any case, safely handling hydrogen and oxygen is necessary as mixtures present an explosive hazard. Often, hydrogen is diluted with air or inert gases (such as nitrogen) and released into the atmosphere to prevent ignition. Alternatively, the hydrogen can be circulated to a fuel cell system to feed power back to the PEMWE, increasing overall system efficiency.²⁸

1.2 Configuration Variations

To demonstrate the versatility of PEMWEs, an overview of their configurations will be presented and the advantages to each. To date, four configurations have seen potential in the literature: conventional operation, high-pressure operation, high-temperature operation, and cathode water feed operation.

Conventional operation is characterized by pressures of ~ 30 bar and temperatures of 50 – 90°C .⁹ Operational pressure is typically defined by hydrogen pressure as the oxygen is usually vented upon generation (and thus is at ambient pressure). Pressure is generated directly from hydrogen gas being produced, assuming the membrane can withstand any associated mechanical stress.²⁹ This so-called “differential pressure” operation provides benefits over isobaric operation because less downstream mechanical compression will be required. However, increased pressure comes greater hydrogen crossover into the anode, resulting in serious safety concerns. Crossover could result in ignitable hydrogen and oxygen gas mixtures on the anode side. Only a 4% volume of hydrogen in oxygen would form an ignitable safety hazard (i.e., 4% is the lower explosive limit or LEL).³⁰ Thus, given current safety limitations, state-of-the-art PEMWEs do not exceed 50 bar. However, as discussed in the next paragraph, there is considerable potential to operate at much higher pressures. Regarding temperature, higher operating temperatures lead to voltage efficiency improvements.³¹ In conventional operation, heat is supplied to the cell and produced by electrochemical losses to maintain a temperature of 50 – 90°C . However, increasing temperatures above the boiling point of water poses a unique set of challenges as 1) state-of-the-art membranes (i.e., PFSA membranes) rely on liquid water-assisted proton conduction, and 2) vapor-fed operation is not well studied.³² Potential strategies to enable high-temperature operation will also be discussed in this section.

High-pressure operation is an attractive alternative to conventional operation as substantial pressurization or liquefaction of hydrogen is required for many end-use cases: 200 bar for gaseous storage, ~ 300 bar for the Haber Bosch process, 350 bar for heavy-duty fuel cell vehicles, and 700 bar for passenger fuel cell vehicles.³³ Since hydrogen is typically produced at 30 bar, adjusting the electrolyzer to output at greater compressions could reduce equipment count, process complexity, capital costs, and maintenance costs, while lowering the energy required to perform final compressive steps.³³ In the literature, high-purity hydrogen at differential pressures up to 350 bar has been shown.³⁴ However, gas crossover problems that already exist in conventional operation are exacerbated under higher pressures.³³ Strategies for mitigating crossover will be discussed in **Section 4.3**.

Higher operating temperatures result in a lower reversible cell potential (e.g., 1.14 V at 200°C versus 1.23 V at 20°C)³⁵ and lower activation overpotentials.³² However, increased component corrosion (e.g., BPs and PTLs) and decreased proton conductivity (due to membrane dehydration) could also result.³¹ Thus, corrosive-resistant materials need to be implemented, and strategies to ensure proton conductivity need to be developed—such as incorporating organic fillers into the membrane (to enhance water uptake)^{36–42} or increasing the water pressure at the anode.³¹

Lastly, novel configurations are also possible and can enable performance improvements. One design by Hystar⁴³ changes the water inlet from anode to cathode and adds air flow to the anode. With these changes, Hystar claims that membrane thickness can be reduced by $\sim 85\%$ (from state-of-the-art $125\mu\text{m}$ to $20\mu\text{m}$) since any crossover hydrogen will be diluted with the anode air feed, mitigating the risk of forming an ignitable gas mixture. Such thin membranes could reduce ohmic losses and increase efficiency substantially, as ohmic overpotentials currently comprise a significant portion of voltage losses ($\sim 40\%$ at a standard operating current density of 2 A cm^{-2}).¹⁸ However, several challenges must be addressed, including removing N_2/CO_2 contaminants in the hydrogen outlet.

1.3 Motivation

PEMWEs will require cost and performance improvements to compete economically against conventional forms of hydrogen production.⁴⁴ The roadmap for GW-scale deployment is complex, but this paper will capture many considerations: deployment targets, government policies, production/capital costs, economies of scale, component-specific improvements, durability testing, and safety regulations. Please note that this paper will focus on cell-level components instead of stack and balance of plant aspects. Two

factors distinguish this review from others: 1) broader economic and political considerations will be discussed along with scientific research goals, and 2) less focus will be put on the current state of PEMWEs, but rather on improvements necessary to reach GW-scale deployment. For reviews on the current state of PEMWEs, we refer the reader to the numerous articles in the literature.^{8,18,45–48}

This paper will be presented as follows: First, global green hydrogen policies will be overviewed to orient the reader to the current hydrogen landscape. Then, targets for PEMWE capital costs and hydrogen production costs will be outlined. Next, component-specific research will be discussed in the context of cost, efficiency, and durability improvements. While research efforts are required for every component, emphasis will be put on the anode catalyst layer, membrane, and PTL. Afterwards, durability testing protocols (i.e., accelerated stress tests) will be proposed for each component. Penultimately, we will zoom out and have three industrial companies—NEL Hydrogen, Plug Power, and an undisclosed company—offer their unique perspectives on how global efforts can be directed for PEMWE expansion. To conclude, all the insights in this paper will be summarized to present a concise roadmap for GW-scale deployment.

2 Deployment Targets

Global hydrogen demand was 94 million tons (Mt) in 2021, accounting for nearly 2.5% of global energy consumption. Nearly half of this demand went towards refining hydrocarbons (46%). The rest was directed to the production of ammonia (35%), methanol (15%), and steel (4%)—by direct reduction of iron.⁴⁹ Transport is a small but growing source of demand, on the order of 30,000 tons (0.03% of global demand), corresponding to a 60% increase over the previous year.⁵⁰ This demand was primarily for fueling trucks, buses, and passenger vehicles, with the potential to expand in the rail, shipping, materials handling, and aviation sectors.

The push to meet hydrogen demand has led to the deployment of MW-scale systems in recent years. Recent deployments include 1) the world-first floating offshore PEMWE in France, utilizing a 1 MW system from Plug Power,⁵¹ and 2) a 1.5 MW load-balancing PEMWE in Germany that feeds hydrogen directly into a natural gas grid.⁵² When considering projects under construction and in the final investment decision (FID) stages, numerous multi-MW systems can be seen.⁵³

2.1 Global Landscape

In recent years, an increasing number of nations have presented hydrogen roadmaps for decarbonization or included hydrogen as part of a clean energy strategy. These roadmaps/strategies often entail plans for hydrogen production/utilization. **Table 1** outlines such policies, ordered by the date of release. This table builds on previous efforts by Wappler et al.⁵⁴ in 2022 by summarizing documents gathered through Australia’s hydrogen policy tracking resource “HyResource.”⁵⁵ In a little over a year since Wappler et al. published their analysis, 26 countries have announced hydrogen policies, increasing the total number of nations to 56.

Table 1. Summary of national hydrogen policies sorted by continent and in chronological order, expanded from ref⁵⁴ and primarily sourced from HyResource.⁵⁵

Country	H ₂ Type	Strategy	Date	Ref	Country	H ₂ Type	Strategy	Date	Ref
Europe					Asia				
Netherlands	Green	Export	Apr-20	56	Japan	Clean	Import	Dec-17	57
Portugal	Clean	Export	May-20	58	South Korea	Clean	Import	Jan-19	59
Germany	Green	Import	Jun-20	60	China	Clean	Export	Mar-21	61
Norway	Clean	Export	Jun-20	62	Singapore	Clean	Import	Oct-22	63
European Union	Green	Import	Jul-20	64	India	Clean	Export	Jan-23	65
France	Clean	Export	Sep-20	66	Sri Lanka	Green	Import	Aug-23	67
Spain	Green	Export	Oct-20	68	Malaysia	Clean	Export	Oct-23	69
Russia	Clean	Export	Oct-20	70	Indonesia	Clean	Import	Dec-23	71

Finland	Green	Export	Nov-20	72	South America				
Italy	Green	Import	Nov-20	73	Chile	Green	Export	Nov-20	74
Ukraine	Clean	Export	Mar-21	75	USA	Clean	Export	Nov-20	76
Hungary	Clean	Export	May-21	77	Canada	Clean	Export	Dec-20	78
Poland	Clean	Export	May-21	79	Paraguay	Green	Export	Jun-21	80
Czech Republic	Clean	Import	Jul-21	81	Colombia	Clean	Import	Aug-21	82
Slovakia	Clean	Import	Jun-21	83	Uruguay	Clean	Export	Jun-22	84
United Kingdom	Clean	Export	Aug-21	85	Panama	Clean	Export	Jul-23	86
Sweden	Green	Export	Dec-21	87	Argentina	Clean	Export	Sep-23	88
Denmark	Green	Export	Jan-22	89	Africa				
Croatia	Green	Export	Mar-22	90	Morocco	Green	Export	Aug-21	91
Austria	Clean	Import	Jun-22	92	South Africa	Clean	Export	Feb-22	93
Belgium	Green	Import	Oct-22	94	Namibia	Green	Export	Nov-22	95
Greece	Green	Import	Nov-22	96	Algeria	Clean	Export	Mar-23	97,98
Luxembourg	Clean	Import	Dec-22	99	Kenya	Green	Export	Sep-23	100
Romania	Green	Import	Jun-23	101	Tunisia	Green	Export	Oct-23	102
Ireland	Green	Export	Jul-23	103	Middle East				
Australia/Oceania					UAE	Green	Export	Nov-21	104,105
New Zealand	Green	Export	Sep-19	106	Oman	Green	Export	Oct-22	107,108
Australia	Clean	Export	Nov-19	109	Israel	Clean	Import	Jan-24	110
					Turkey	Clean	Export	Jan-23	111

For hydrogen production there are a several distinctions drawn by method of production, and often a distinction drawn between “green” and “clean” hydrogen. Clean hydrogen is a more general term to refer to processes that result in relatively little or no carbon emissions. Clean hydrogen includes green hydrogen but also includes processes such as “blue” hydrogen, where hydrogen is produced via steam methane reformation with carbon capture and underground storage (CCUS). Blue hydrogen (and in some cases, grey hydrogen—or hydrogen directly produced from SMR without CCUS) is presented as a bridging device¹¹² to meet short-term demands for hydrogen while green hydrogen technologies gradually become online. Short-term demands of clean and green hydrogen include the steel industry of the European Union¹¹³ as well as transport fuels to support the development of a green hydrogen transport fuel market. In this vein, certification schemes for tracking the greenhouse gas emission intensity of hydrogen (e.g., distinguishing between clean, green, blue, grey, and other forms of hydrogen) are under development, such as with the European Union’s CertifHy¹¹⁴ and Australia’s Guarantee of Origin (GO) scheme.¹¹⁵

When considering the production of green hydrogen from a life cycle analysis (LCA) standpoint, water electrolysis entails the lowest natural resource costs, human health impacts, and ecological impacts, when combined with wind or nuclear power (as opposed to solar photovoltaics, due to their relatively intense environmental impact from a life cycle standpoint).^{116–118} When considering external cost factors (human health impacts, ecological impacts, resource depletion), electrolysis coupled with wind-power is one of the most cost effective methods of hydrogen production, green or otherwise.¹¹⁷ Lastly, in terms of global warming potential, water electrolysis is slightly more carbon intensive than biomass gasification with carbon capture (which has the potential to reach net negative CO₂ equivalent emissions), although the latter entails far greater emissions of criteria pollutants and thus has higher externalized cost factors over its lifetime.^{117,119}

2.2 2030 Targets

Figure 2 displays stated 2030 targets for electrolyzer capacity, sourced from national policy documents in **Table 1**. The goals are expressed in equivalent nominal electrolyzer power capacity. When annual green hydrogen production rates were given instead of capacity, the IEA convention of 50 kWh/kg H₂ and an

assumed annual capacity factor of 50% was used to estimate the equivalent capacity.¹²⁰ While many European countries have released some form of plan involving electrolysis and/or green hydrogen production targets, there is a fair amount of variation in approach with the Netherlands, Norway, France, Ukraine, Hungary, Poland, Czech Republic and the United Kingdom all emphasizing some amount of clean H₂ production rather than entirely green, as in the case of Portugal, Germany, Spain, Finland, and Italy. In Asia, India and China have also announced dual strategies of green and clean hydrogen production, with green hydrogen amounting to 5 Mt/yr for India and 1 Mt/yr for China. In South America, Chile, Colombia and Uruguay seek to leverage their abundant renewable energy resources, including hydropower,^{121,122} to become net exporters of green hydrogen, with Chile targeting the installation of 25 GW of electrolysis by 2030. Several countries in Africa and the Middle East are scaling up green hydrogen production to be large exporters of green hydrogen as well. Countries in these regions are also planning to sustainably produce commodities that are traditionally associated with greenhouse gas emissions, with Egypt¹²³, Namibia¹²⁴, and the United Arab Emirates^{105,125} to export green ammonia, and Morocco to export green methanol and other liquid fuels.^{126,127}

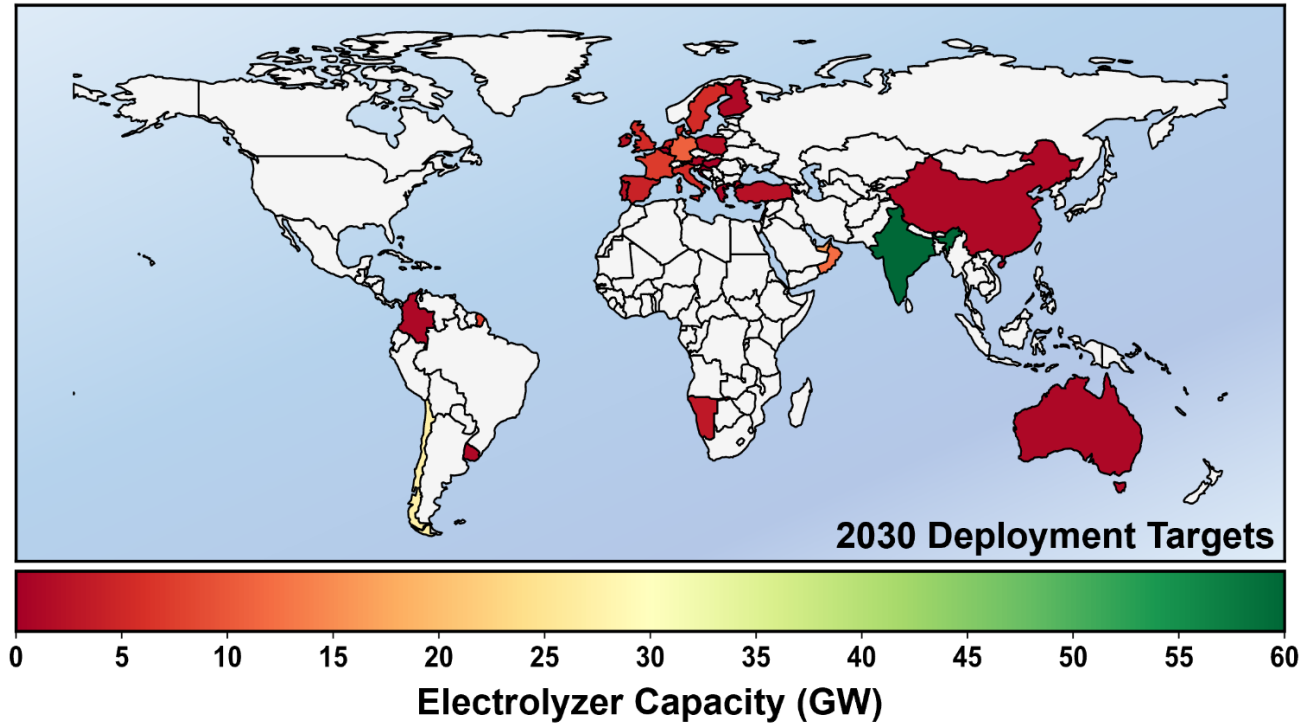
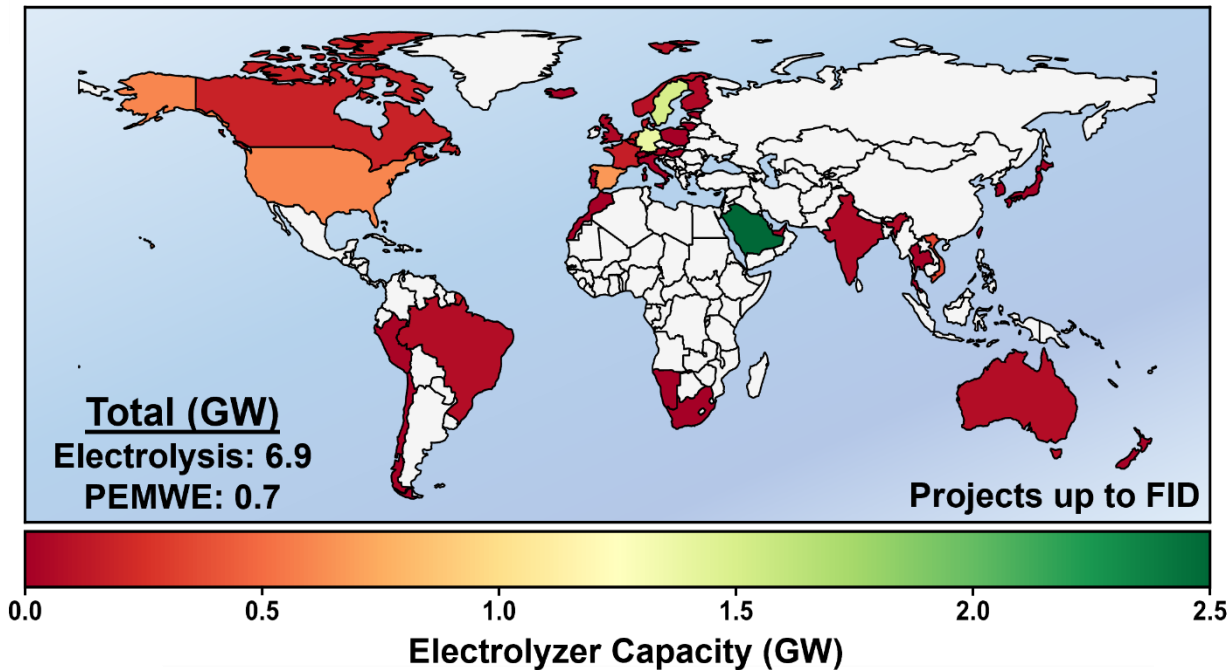


Figure 2. Stated 2030 targets for electrolyzer capacity, sourced from documents in **Table 1**. Targets are expressed in equivalent nominal electrolyzer power capacity (GW); for documents that used hydrogen production as the metric, the IEA convention of 50 kWh/kg H₂ and an assumed annual capacity factor of 50% was used to estimate the equivalent nominal electrolyzer capacity.

2.3 Active vs Planned Projects

Using the IEA database, electrolyzer deployment projects based on nameplate capacity are examined in

(a)



(b)

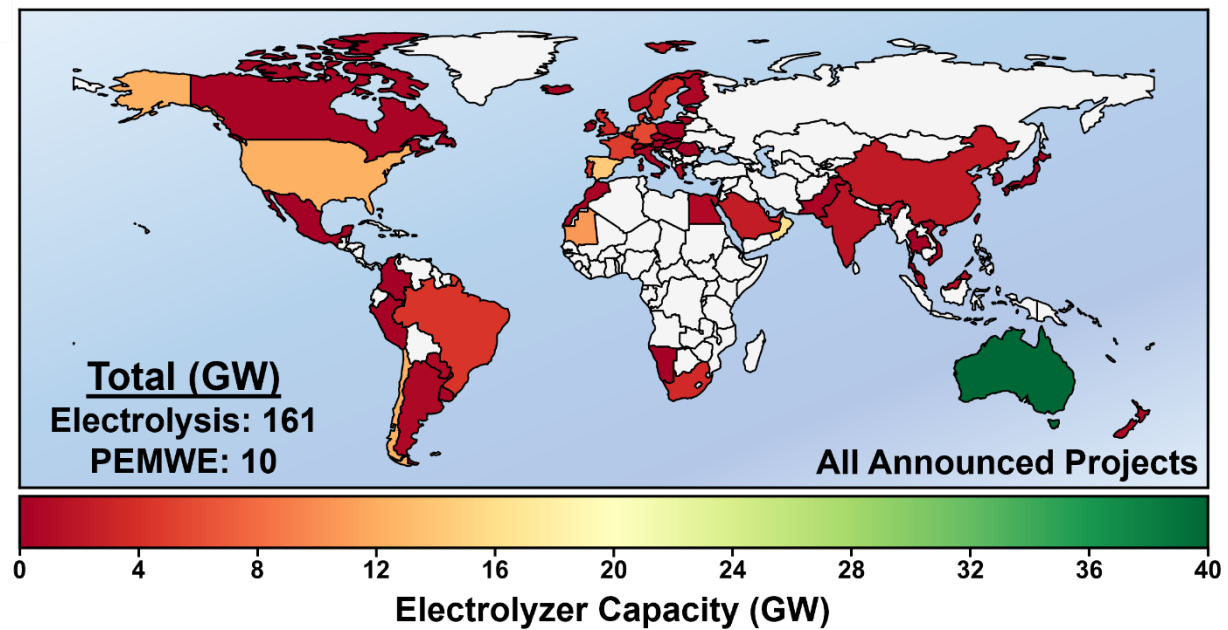
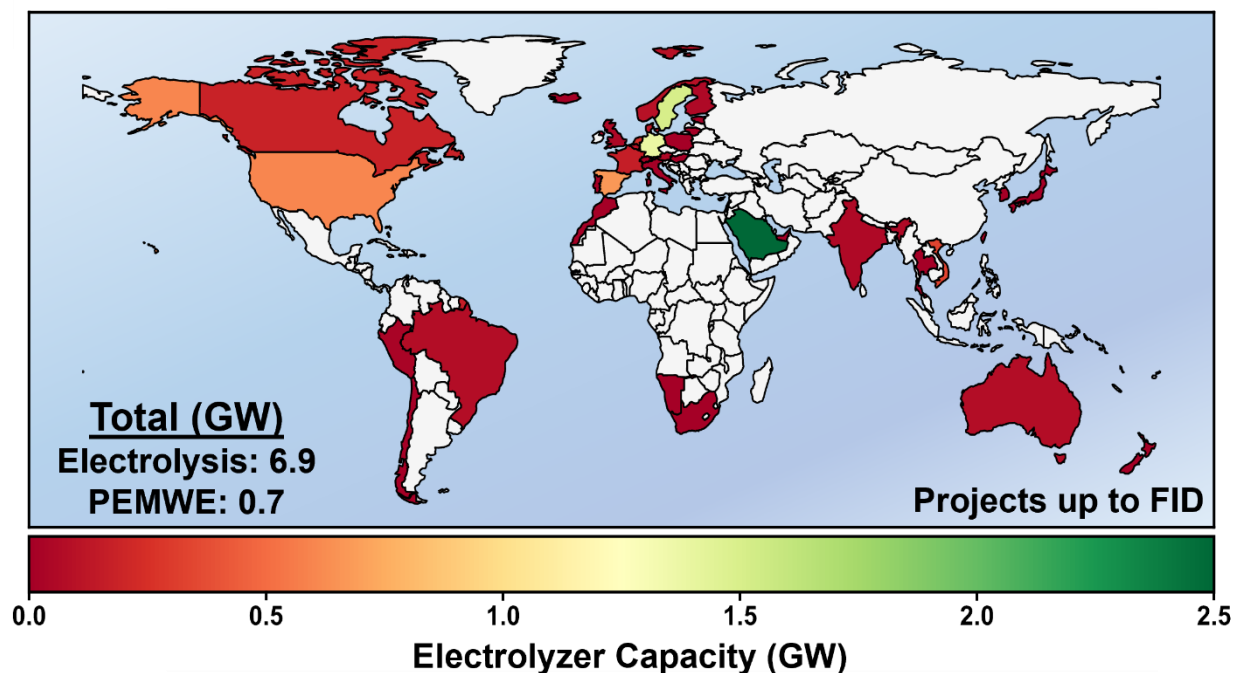


Figure 3, with distinction drawn between currently active (or nearly active) and planned projects.¹²⁰

(a)



(b)

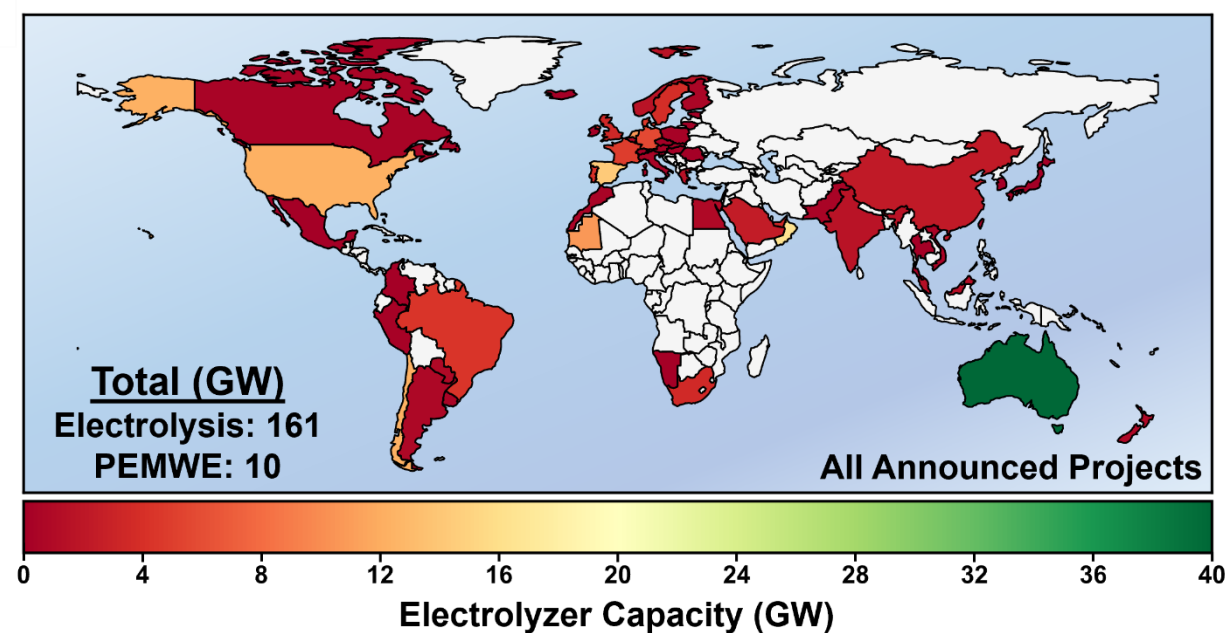


Figure 3a depicts electrolyzer deployment projects that are currently active or will be active in the near future, which include projects that are actively operating, under construction, and up to final investment

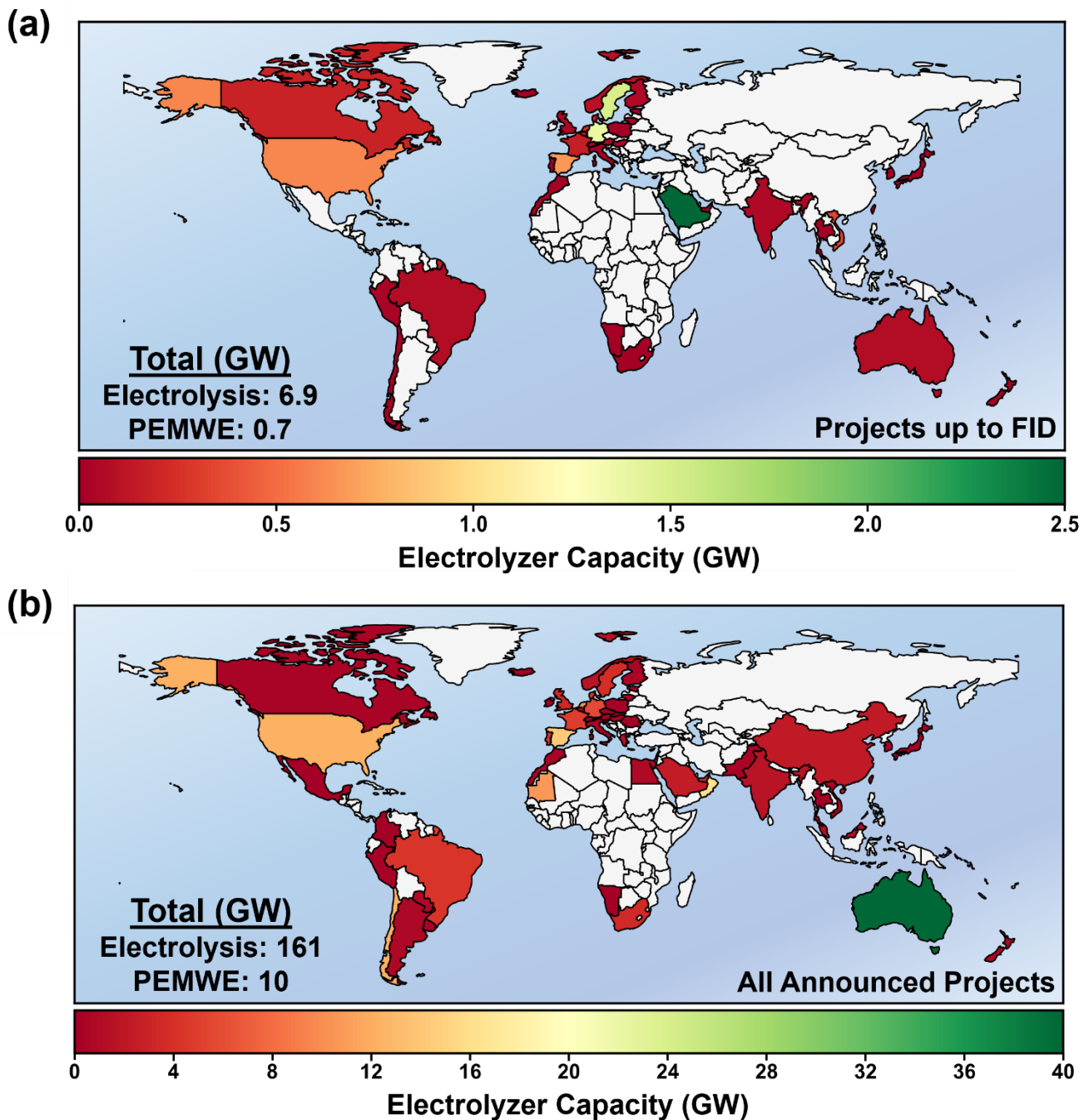


Figure 3b shows projects that are announced but still in early planning stages, i.e., projects that are not up to FID. Between the two scenarios is a large discrepancy in total electrolysis deployment, with 7 GW up to FID, amounting to less than 5% of the 161 GW of announced targets. For PEMWEs, there is 0.3 GW currently in operation with another ~0.4 GW in construction or having received FID, and a further ~10 GW in the early planning stages. Projections in expected PEMWE deployment to meet decarbonization goals set by IEA and the Hydrogen council were found to require between ~80 to 220 GW of PEMWE installed capacity by 2030, assuming a 40% market share with other electrolysis technologies.⁴ Clearly, there is a need to rapidly accelerate the global deployment rate to reach such targets.

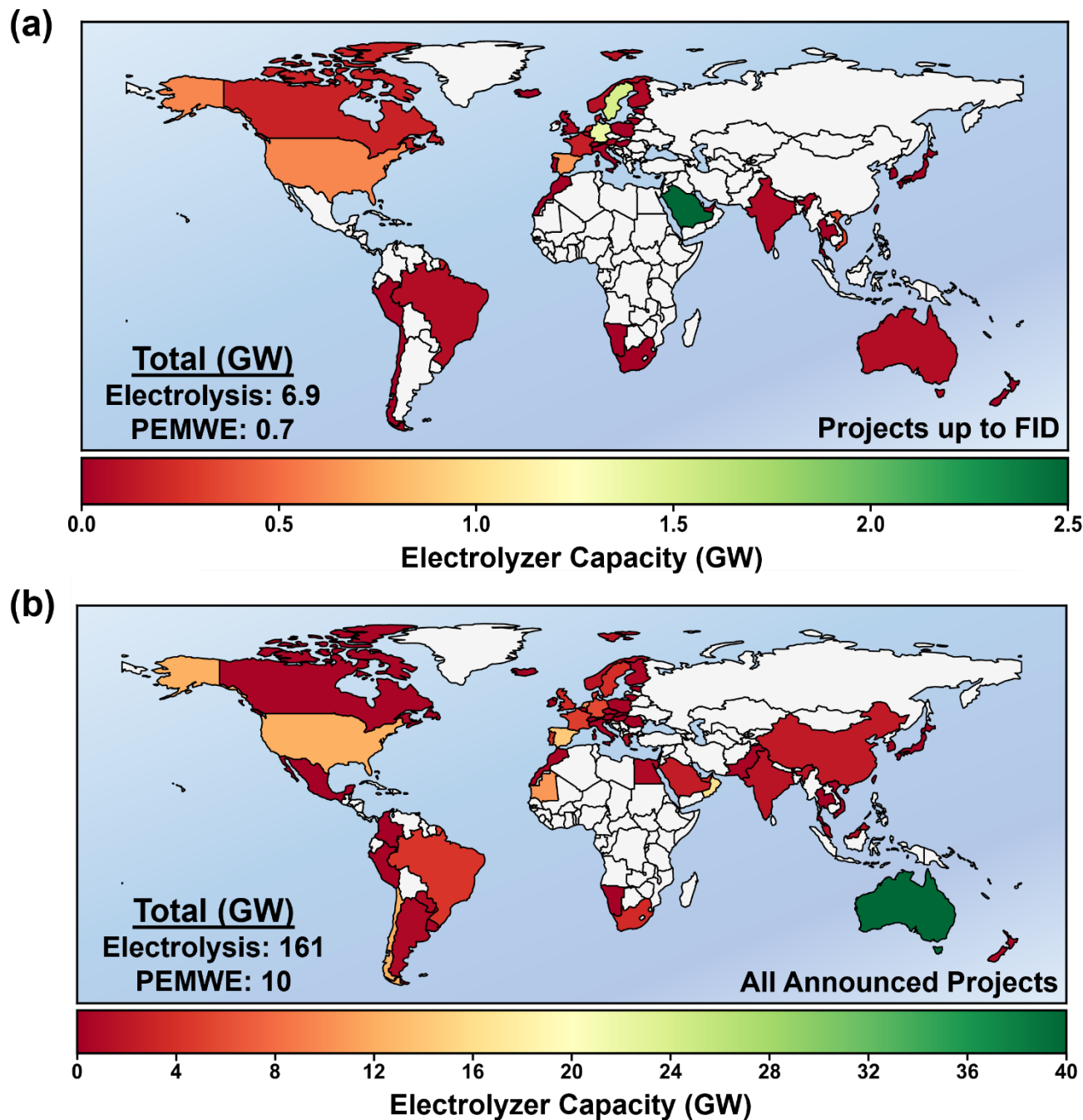


Figure 3. (a) Electrolyzer projects that are active, undergoing construction, or have announced FID. (b) All announced electrolyzer projects, including active ones. All projects sourced from IEA H₂ projects database.¹²⁰

Notably, policy targets alone are not fully indicative of the electrolysis deployment landscape. While countries such as the United States, Japan, and Saudi Arabia do not have targets that specify green hydrogen production or electrolysis deployment, there are still appreciable amounts of active electrolyzer systems with further deployments expected. In some cases, national targets are outstripped by planned deployments; the Australian government has announced goals of 1 GW of electrolyzer capacity by 2030, however there is already 11 GW of projects in various stages of planning. Policy targets themselves continue to be moving targets as well, largely trending upwards in hydrogen production targets with each revision.

3 Cost Targets

The United States Department of Energy (US DoE) announced cost targets of \$2/kg H₂ by 2026 and \$1/kg H₂ by 2031 for clean hydrogen. In 2021, hydrogen production costs ranged from \$1.0-2.5/kg H₂ for steam methane reformation (SMR), \$1.5-3.0/kg H₂ for SMR with carbon capture and \$4.0-9.0/kg H₂ for electrolysis.⁴⁹ A multitude of factors—including gas supply constraints, energy security concerns ignited by the invasion of Ukraine, and the exiting of a global pandemic—led to surging natural gas demand and, consequently, prices as high as \$85-153/MWh around much of Europe and significant parts of Asia after holding steady in a range of ~\$10-23/MWh since 2015.¹²⁸ Thus, for the affected regions, hydrogen production costs (via SMR without carbon capture) skyrocketed to \$4.8-7.8/kg H₂ in Europe and \$5.3-8.6/kg H₂ in Asia, equating to \$121-198/MWh and \$134-218/MWh, respectively.¹²⁹ While natural gas prices may decline in the next few years to 2015 levels, and indeed have already returned to pre-pandemic levels across much of the globe, such volatility highlights the potential for cost-competitive electrolytic hydrogen powered by diverse and potentially more reliable renewable resources. Furthermore, SMR production of hydrogen is highly sensitive to the cost of natural gas. Without carbon capture, SMR consumes approximately 0.039–0.055 MWh_{nat gas}/kg H₂, while SMR with carbon capture consumes 0.077 MWh_{nat gas}/kg H₂.^{119,130} Thus, during volatile natural gas price swings—as recently experienced in Asia and Europe—the price of hydrogen from SMR can increase substantially; for example, if natural gas prices rise from \$20/MWh (2015 levels) to \$100/MWh (2022 levels), the cost of SMR-produced hydrogen would increase \$3.1-6.2/kg H₂ (depending on whether carbon capture is used), quickly closing the price gap between SMR and electrolysis.

3.1 Cost Factors

3.1.1 Electricity Costs

The ability for electrolytic hydrogen to become cost competitive is strongly dependent upon the availability of low-cost electricity.¹³¹ However, even with relatively cheap electricity prices of \$0.05-0.06/kWh, the cost of the electricity alone to make electrolytic hydrogen would equate to \$3/kg (with no consideration for capital or other operating expenditures), making it difficult to compete with SMR (with *total* costs being \$1.0-2.5/kg H₂).¹³² Nonetheless, thanks to the increasing availability of lower cost renewable electricity in the form of solar and wind power, electricity prices equivalent to \$0.02/kWh have been observed, which would enable \$1/kg H₂ in electricity costs.¹³³ To meet a competitive cost point for electrolytic hydrogen, capital expenditures (CapEx) of electrolysis—defined as the cost of the electrolyzer and ancillary equipment, along with installation and other fixed costs—would also need to meet an equivalent of \$1/kg H₂ whilst utilizing this low-cost electricity. This may be currently feasible with high capacity factors (defined as the % of system utilization), but for pairing electrolysis with renewable energy sources that provide low-cost electricity, capacity factors on the order of 30-40% are more realistic.^{134,135}

3.1.2 Renewable Energy Coupling

Increasing capacity factor is the second most important cost factor, behind only electricity costs (**Figure 4**).¹³⁶ However, to couple PEMWEs with renewable energy, periods of low capacity factors will be inevitable. Thus, the temptation is to constantly have PEMWEs draw power from the grid, even during such periods. However, coordinating with the grid to provide energy during these times is not always possible and is generally prohibitively expensive. Furthermore, even if there is enough power locally, utilities prefer PEMWEs only to draw power during periods of excess renewable supply so as not to exasperate grid balancing issues. Thus, grid control algorithms and management strategies are required.

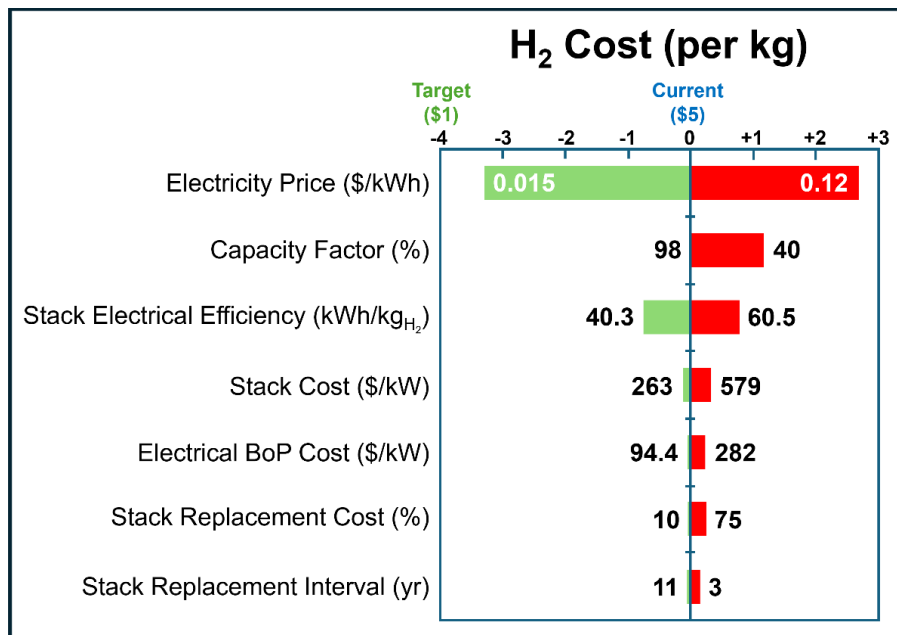


Figure 4. Sensitivity analysis for cost factors of hydrogen. Adapted from ref.¹³⁶ Copyright 2020 U.S. Department of Energy.

Since current PEMWEs are optimized for constant load operation, load fluctuations from renewable energy coupling can also accelerate the degradation of critical components, leading to lower system lifetimes. For example, anode CLs are especially sensitive to such fluctuations and can be the limiting factor in stack lifetimes.¹³⁷ BoP components (e.g., pumps, driers, and hydrogen compressors) can also suffer from intermittent operation. Since system lifetimes directly influence overall H₂ costs, innovative strategies to provide more consistent power—such as utilizing energy storage, dynamic controls, or multiple renewable sources—are necessary. Lastly, degradation can be minimized via intelligent control strategies for larger systems. An example can be demonstrated using a 600 MW plant

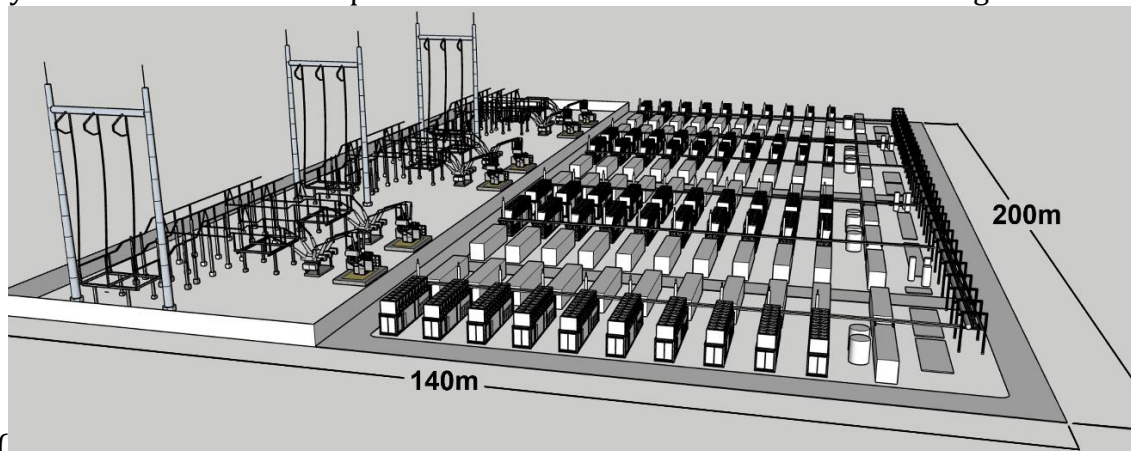


Figure 5) consisting of sixty 10 MW arrays (arrays contain multiple stacks): if the power input for a given period oscillates from 100-300 MW, instead of operating all sixty arrays at 17-50% power, one can operate thirty arrays at 33-100% power.

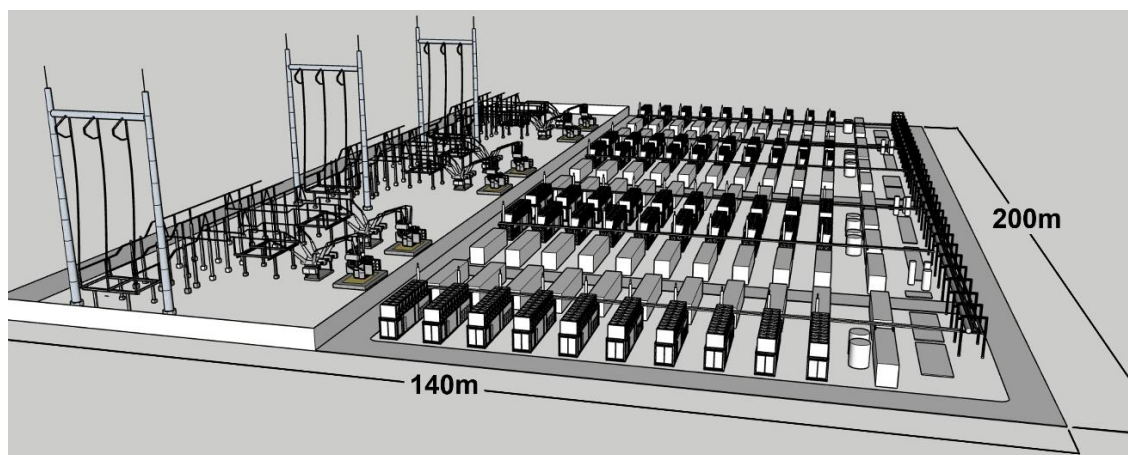


Figure 5. 600 MW PEMWE concept plant utilizing sixty 10 MW arrays (from Plug Power).

In summary, balancing renewable energy integration with potential economic repercussions—namely, low capacity factors and increased degradation—will be essential as PEMWEs scale up. However, given the limitations of electricity costs and renewable energy coupling, reductions in CapEx will be necessary to meet hydrogen cost targets.

3.1.3 Capital Expenditures

Given its hydrogen cost targets (\$2/kg by 2026 and \$1/kg by 2031), the US DoE has carried out an analysis that indicates that PEMWE CapEx of \$300/kW by 2026 and \$150/kW by 2031 are required.¹³⁸ Assuming a modest 50,000 hours of operation, a PEMWE produces approximately 1000 kg H₂/kW nameplate capacity, and thus a CapEx of \$150/kW would contribute \$0.15/kg $\left(\frac{\$150}{\text{kW}} \times \frac{\text{kW}}{1000 \text{ kg}} \right)$ to the H₂ production cost. Further studies indicates that even with such capital cost reduction in a short period of time, the latter target of \$1/kg H₂ is still out of reach for electrolytic hydrogen due to operating expenditures (OpEx) related to the price of electricity, even in regions of the United States with high renewable electricity potential. Thus, further reductions in capital costs than those stated by the DoE are required for both PEMWE and renewable electricity sources.¹³⁹ The OpEx of electrolytic hydrogen can be reduced by R&D towards more efficient or durable PEMWEs and by operating in lower current density regimes (although the latter entails a trade-off with increasing CapEx due to low capacity factors, leading to variable optimal current density regimes as a function of electricity costs).¹³

Current CapEx of PEMWEs are observed in a wide range of around \$800-1500/kW.¹⁴⁰⁻¹⁴⁴ Future capital costs associated with PEMWE have been estimated in the literature by several methods including literature surveys,^{131,145} industry surveys,^{146,147} learning curve estimates,^{144,148} and bottom-up cost analyses.^{142,149} Across the breadth of these studies, future capital costs of PEMWE are generally expected to hit the range of \$200-600/kW, with high variation largely due to differences in assumptions regarding 1) the rate of production scale-up, and 2) the impacts of economies of scale and learning rates on cost reduction.¹⁵⁰ **Figure 6** shows several recent cost estimate projections using a variety of techno-economic (TEA) methods, along with notable cost targets based on decarbonization goals. Cost targets clearly hinge on optimistic CapEx reduction scenarios or lie outside of current projections. Considerable research and development are required to bridge this gap between CapEx targets and current cost projections.

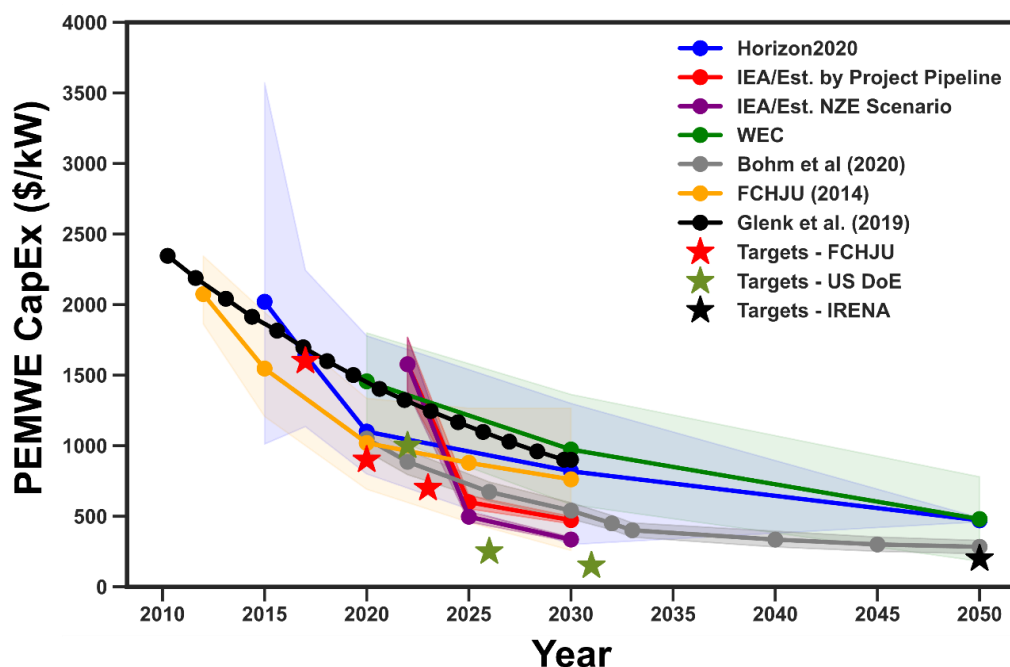


Figure 6. Comparison of several projections in the literature for expected CapEx of PEMWE systems with relevant confidence intervals shaded in (Horizon 2020¹⁵¹, IEA⁴⁹, WEC¹⁵², Bohm et al.¹⁴⁴, FCHJU¹⁴⁶, Glenk et al.¹³¹) as well as cost targets set for enabling PEMWE as a competitive technology for hydrogen production (FCHJU¹⁵³, US DoE¹⁵⁴, IRENA¹⁴¹).

3.1.4 Carbon Taxes

Before discussing methods to reduce CapEx, it should be noted that placing a price on carbon emissions for hydrogen production—either in the form of carbon credits or carbon taxes—can be a significant economic driver for green hydrogen. For PEMWEs to reach cost parity with SMR-based H₂ production, a carbon price of \$230-550/ton CO_{2e} (“e” is global warming potential equivalent) is required. SMR with carbon capture is expected to reach cost parity with SMR (no carbon capture) with a more modest \$50-60/ton CO_{2e} cost of carbon.⁵³ Carbon markets have grown tremendously in recent years, with 24% of global emissions covered by some form of a national carbon pricing program. Notably, emissions trading systems (ETS) such as the E.U. ETS and California’s Cap and Trade Program experienced prices of \$60/ton CO_{2e} and \$40/ton CO_{2e} for the year of 2023.

3.2 CapEx Reduction

The primary driver to decreasing CapEx for PEMWE over the next decade is increasing manufacturing capabilities in both unit size and production volume, with companies aiming for MW-scale unit sizes (compared to kW-scale) and GW/yr production capacities. In addition to improving equipment utilization and spreading fixed costs (such as labor) over a greater production capacity, these cost reductions also entail implementing more effective manufacturing, which include automation and roll-to-roll assembly.¹⁵⁵

PEMWE systems consist of the stack and the balance of plant (BoP), which provides the fluids handling, thermal conditioning, power conditioning, and controls needed to facilitate the reactions occurring in the cells. According to a detailed TEA of CapEx as a function of stack size (**Figure 7**), a cost split of roughly 40% stack and 60% BoP is observed at lower manufacturing volumes for 1 MW systems,¹⁴⁹ which is generally in-line with other analyses.^{134,141,142} As production rates scale-up, the BoP expenditures becomes increasingly dominant, comprising upwards of 70% of the manufacturing costs. A third of the manufacturing costs can be attributed to power supplies alone, which is required to convert high voltage AC or DC input sources to the low voltage and high current DC electricity needed for electrolysis. AC/DC thyristor-type rectifiers are most commonly utilized due to their reliability, technological maturity, and comparatively low cost.¹⁵⁶ The effect of economies of scale is presumed to not be as impactful on balance of plant components, which are largely outsourced. This was observed to be the case with BoP costs for

PEM fuel cell (PEMFC) power plants as they scaled up, particularly with the AC/DC power conditioning equipment.¹⁵⁷

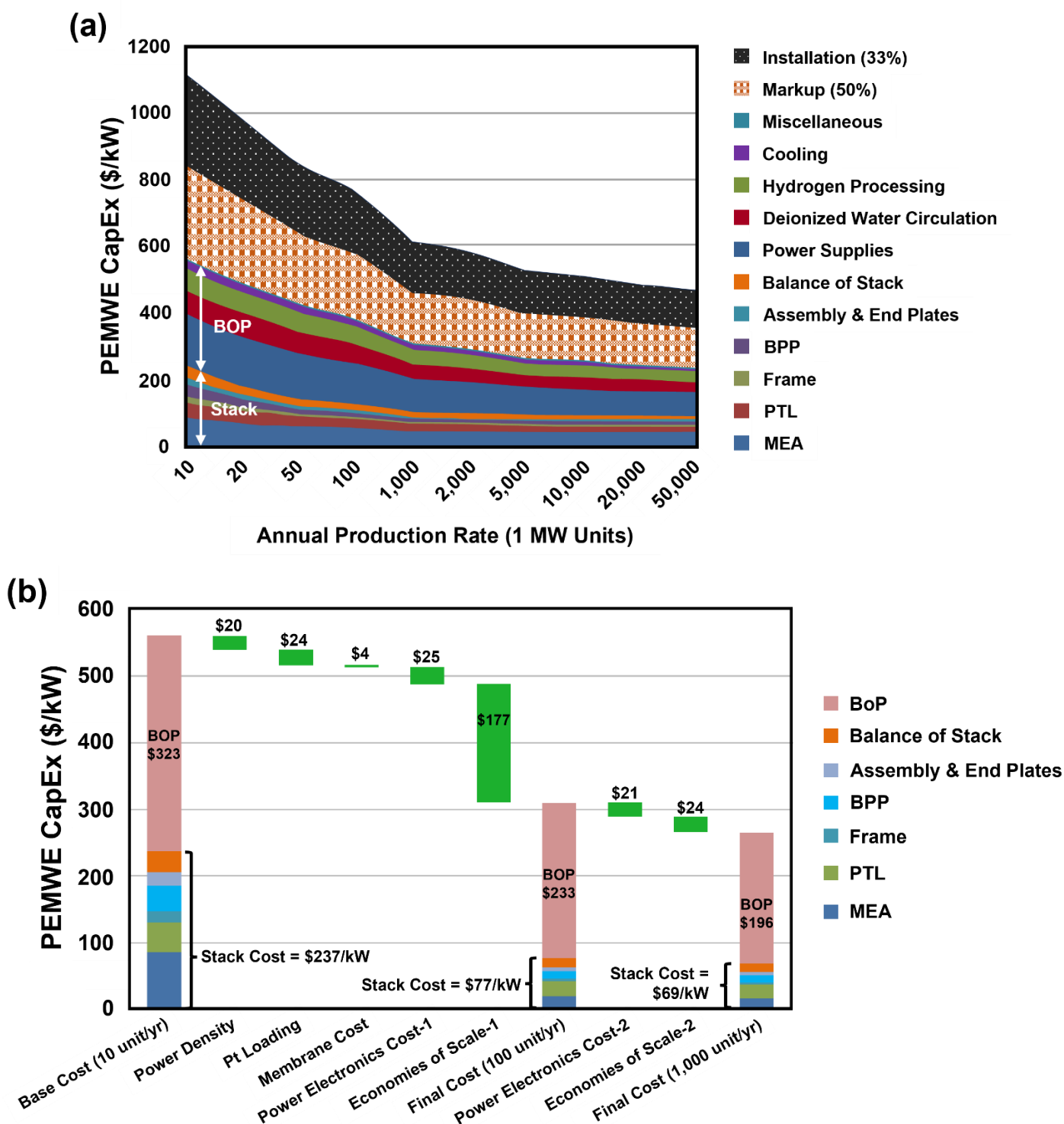


Figure 7. Capital cost of 1 MW PEMWE system breakdown by bottom-up manufacturing analysis, projected out by production volume (a) and potential for R&D driven cost reductions of 1 MW PEMWE systems (b). “Economies of Scale” encompasses manufacturing engineering for (b). Adapted from ref.¹⁴⁹ Copyright 2019 National Energy Renewable Laboratory (NREL).

At the stack level, cost can be broken down into CCMs, PTLs, GDLs, BPs, end plates, frames, balance of stack, and assembly labor. Balance of stack refers to additional components such as wires, insulation, and compression hardware and can make up around 13% of the stack cost.¹⁵⁸ In order of contribution,

CCMs are the most expensive component, followed by PTLs, BPs, and then end plates and framing. At higher production volumes where labor costs are lower, material costs begin to dominate and MEAs can comprise nearly half the stack cost due to the expensive membrane material and the platinum group metal (PGM) catalyst used. PTLs are constructed out of titanium and bipolar plates are most commonly stainless steel, both of which require coatings that are typically gold or platinum.

In order to reach the U.S. DoE CapEx target of \$150/kW by 2031, stack production costs need to be brought down significantly from current values of \$237/kW. Several pathways have been identified to reduce stack costs, primarily focused on reducing material costs, increasing system efficiency, or achieving higher current density capabilities.¹⁵⁹ Although current densities as high as 15 A·cm⁻² have been observed,¹⁶⁰ obtaining greater current densities alone is not sufficient to reduce costs as such high current operation is only beneficial when low cost electricity is available.^{161,162} Thus, focus needs to also be put on reducing material costs and improving stack efficiency, both of which will require the optimization of individual components at the cell level.

4 Component Improvements

This section will discuss all the primary components in a PEMWE cell and establish goals for rapid deployment. Challenges in fundamental research and large-scale considerations—such as cost and manufacturability—will be thoroughly reviewed.

4.1 Anode Catalyst Layer

A conventional anode catalyst layer (CL) consists of a catalyst dispersed in an ionomer (an ionically conductive polymer) network coated on either a membrane—in the case of a catalyst-coated membrane (CCM) configuration—or a PTL—in the case of porous transport electrode (PTE) configuration. Additionally, membrane electrode assembly (MEA) is a general term that collectively refers to the membrane and adjacent CLs, with no distinction between CCM and PTE configurations. Multiple methods to coat a CL onto a substrate (e.g., a membrane or PTL) can be employed, such as airbrushing, ultrasonic spraying, or doctor blade coating.¹⁶³ The MEA configuration and CL fabrication method can influence CL properties; such impacts have been thoroughly discussed in a review by Wang et al.¹⁶³ Thus, this section will focus on smaller-scale considerations, such as loading reductions, catalyst utilization, catalyst supports, and CL microstructure.

The catalyst must be stable under the highly acidic (pH~0) and oxidizing (>1.3V) conditions of the anode, which currently limits the material selection to precious metals.¹⁶⁴ While ruthenium is one of the most active OER catalysts, its lack of stability has resulted in iridium being the primary choice.⁸ Iridium oxide (IrO_x) is the most commonly used form since metallic iridium is less active.^{165,166} Activity and durability are highly dependent on the surface state of the iridium, with amorphous IrO_x showing higher activity and lower durability than crystalline IrO_x.¹⁶⁷ Mom et al.¹⁶⁷ attributed these properties of amorphous IrO_x to greater deprotonation depth under high potentials. However, the catalyst can undergo chemical changes when experiencing varying cell potentials or being in contact with H₂ (from cathode crossover). Rheinlander et al.¹⁶⁸ reported that crystalline IrO₂ can be transformed to amorphous IrO_x or even metallic Ir (based on operating conditions). Thus, changes in the catalyst state should be meticulously considered, especially during long-term testing.

Minimizing the amount of iridium used in anode CLs is vital for reaching cost targets. Between the use of platinum in the cathode and iridium in the anode, iridium is the bottleneck in scale-up due to its rarity (7.5 tons of iridium is mined per year, compared to ~200 for platinum¹⁶⁹).¹⁷⁰ In the literature, notable progress has been made to reduce loadings from >1 mg_{Ir}·cm⁻² to below 0.5 mg_{Ir}·cm⁻².^{171–177} However, further efforts must be made to enable large-scale fabrication of these low-loading CLs, such as 1) obtaining a fundamental understanding of interfacial transport mechanisms, 2) identifying degradation modes, and 3) ensuring quality control at greater production capacities.¹⁶³ Understanding transport mechanisms by unveiling the interplay between electrons, protons, water, and oxygen in low-loading CLs is required to achieve comparable performance with high-loading CLs. Moreover, identifying degradation

modes will enable us to tune CL structure for improved durability. Lastly, translating laboratory-scale fabrication methods to large-scale production for low-loading CLs is of utmost importance for scale-up but has largely been overlooked in the field.

4.1.1 Catalyst Utilization

Optimizing transport processes in the CL (illustrated in **Figure 8**) is a prerequisite for high catalyst utilization at low loadings. To sustain the OER, the CL must facilitate the transport of protons, electrons, liquid water, and gaseous oxygen. Protons move through the ionomer phase in the CL to the membrane. Electrons are transported through iridium particles in the CL to the grains in the PTL. Notably, isolated iridium particles and particles that are fully embedded in the ionomer phase will likely not conduct electrons as ionomers are electronically insulating. Lastly, the PTL allows liquid water to permeate through its pores (in the scale of tens of micrometers)¹⁷⁸ to the pores in the CL (in the scale of nanometers)¹⁷⁹ while simultaneously facilitating oxygen gas removal.

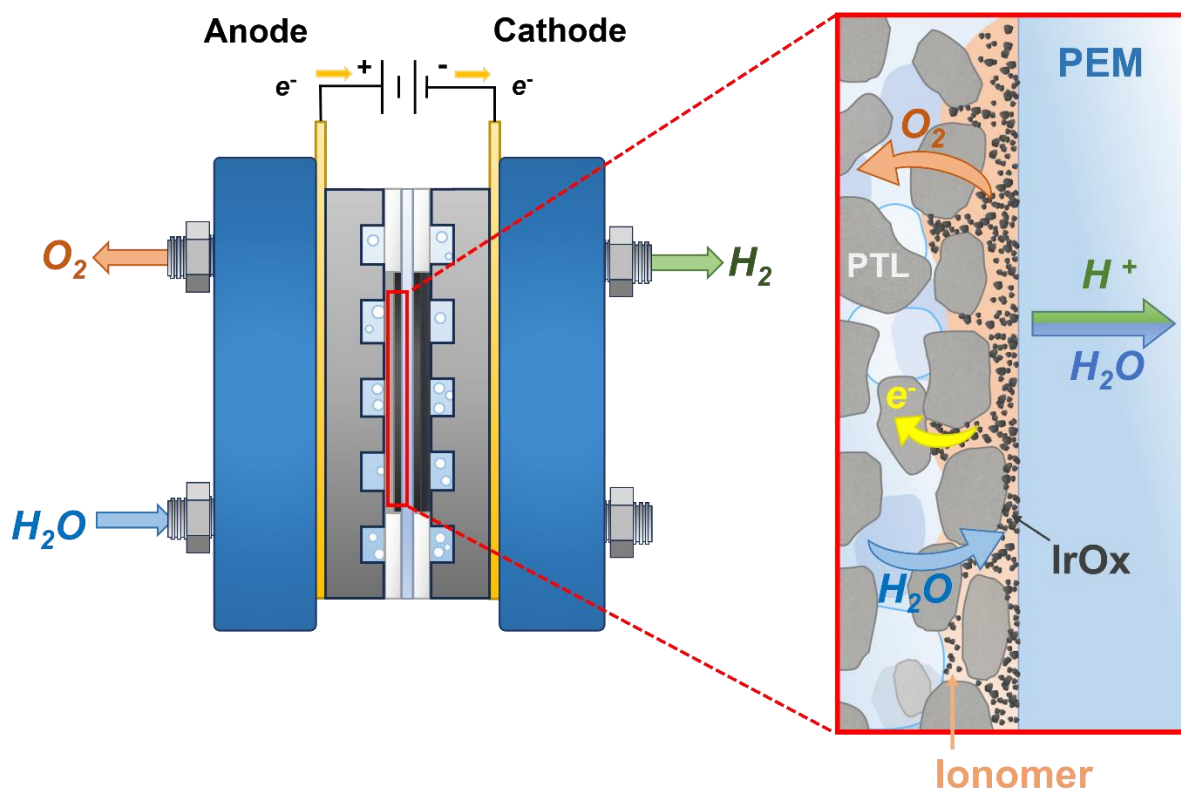


Figure 8. Transport processes occurring at the CL-PTL interface. Reactant liquid water transports from PTL pores to CL pores. Product gases flow in the counter direction: from CL reaction sites to PTL pores. Ionic transport occurs through the ionomer phase and the membrane, while electrical transport occurs through the iridium in the CL and particles in the PTL.

The interface where PTL, catalyst, and ionomer meet—termed the “triple-phase boundary”—plays a vital role in catalyst utilization as the OER can only occur here. Notably, iridium particles that do not directly contact the PTL can still participate in the OER if they are electrically connected to the PTL through other particles. At low iridium loadings, such a connected catalyst network is less likely to exist. Thus, particles not directly contacting the PTL are less likely to be utilized, reducing the triple-phase boundary. Consequently, when CLs swell (simply due to water uptake from the ionomer) into the void space of the PTL interface—known as delamination—the iridium particles in the CL spread apart and become more disconnected, leading to lower electronic conduction and, consequently, low catalyst utilization.¹⁸⁰ At low loadings, poor catalyst utilization resulting from delamination is particularly severe due to the low content of iridium particles.

While low iridium loading MEAs have shown high performance in some cases,¹⁶³ strategies to alleviate issues of electronically insulated—and thus unutilized—iridium particles will likely be required. Due to the dependence of catalyst utilization on both the CL and PTL, strategies to improve utilization regarding the PTL will be discussed in **Section 4.4.4**. However, strategies for the CL also exist. The first is proposed by Hegge et al.,¹⁸¹ where isolated iridium particles are bridged together by depositing a layer of iridium fibers (fabricated via electrospinning) on top of them. The second is by Chatterjee et al.,¹⁸² where utilization was enhanced by completely changing the catalyst morphology: instead of iridium nanoparticles, the authors synthesized nanoporous iridium nanosheets (via electrochemical dealloying of nickel-iridium foils) that provide a continuous, connected structure. While these two methods are more novel, utilizing catalyst supports to increase utilization has been widely studied.

4.1.2 Catalyst Supports

In principle, dispersing a catalyst on a support should increase its surface area to mass ratio, reducing the catalyst mass needed to achieve a desired current. Thus, supports hold significant potential for loading reductions. As detailed below, the critical challenge is that the most commercially available support, TiO₂, is electrically insulating; thus, the benefit of increased catalyst dispersion is partially countered by electrically isolating the catalyst. Many research efforts have long sought to develop conductive support materials, either by doping TiO₂ or finding new materials entirely, but none have achieved long-duration durability testing as TiO₂ has. If TiO₂ is used, the iridium oxide must act as the electron conduit (not the TiO₂). Therefore, connected pathways of iridium oxide on the catalyst support and within the CL are needed to ensure iridium utilization.

4.1.2.1 Material Selection

For supports to be viable, the material must be stable in the operating conditions. The highly acidic (due to the presence of protons from the OER) and oxidizing conditions of the anode render most support materials—conductive carbon black included—thermodynamically unstable.¹⁸³ Most of the remaining (more) stable materials are semiconducting metal oxides, which present inherent conductivity concerns. Nonetheless, several novel catalyst supports have demonstrated promising performance in rotating disk electrode and/or MEA setups, including TaC,^{184,185} Sb₂O₅-SbO₂ (ATO),^{186–188} NbC,¹⁸⁵ TiC,¹⁸⁵ WC,¹⁸⁵ NbO₂,¹⁸⁵ and W₂B.¹⁸⁹ Their accomplishments notwithstanding, this review will not cover these supports given the scarce literature with low loadings and stability testing at industrially relevant current densities (greater than 1 A·cm⁻²). We refer the reader to a review on emerging supports,¹⁹⁰ and we look forward to more progress in novel designs.

TiO₂ as a support has increasingly been tested at industrially relevant current densities with low iridium loadings and thus will be the focus of this section. The interest in TiO₂ as a support for the OER can be traced back to the commercial implementation of RuO₂ on TiO₂ via dimensionally stable anodes (DSAs®) for the chlorine evolution reaction, which has an even higher potential than OER (1.358 V vs SHE).¹⁹¹ This technology established the relevance of metal oxides as anode electrocatalysts and supports. There are primarily two commercial IrO_x/TiO₂ (i.e., IrO_x supported on TiO₂) catalysts in recent literature: the Umicore Elyst Ir75 (IrO₂/TiO₂, diameter ~28 nm)^{168,192–197} and the Heraeus P2X (IrOOH_x/TiO₂, diameter ~284 nm).^{194,195,197}

4.1.2.2 Resistivity Issues

Despite its high stability, TiO₂ supports come with a caveat: high electronic resistivity. While a common catalyst support for PEMFCs (Vulcan-XC72) has a resistivity of $\sim 0.4 \Omega \cdot \text{m}$, values for TiO₂ (in nanowire form) are upwards of $4000 \Omega \cdot \text{m}$.¹⁹⁸ Several studies have attempted to bypass this issue by using more reduced forms of titanium. At low loadings ($< 0.3 \text{ mg}_{\text{Ir}} \cdot \text{cm}^{-2}$), IrO₂/Ti has been shown to increase ECSA and overall outperform unsupported IrO₂.¹⁹⁹ Others have studied Magneli phase titanium oxides (i.e., substoichiometric oxides of the form Ti_nO_{2n-1}), finding increased electronic conductivity relative to TiO₂²⁰⁰ and enhanced performance (using a PTE configuration) relative to unsupported MEAs.²⁰¹ However, whether these supports remained in their reduced state throughout operation was left unaddressed.

Since IrO₂ films and single crystals have resistivity values multiple orders of magnitude lower than TiO₂ ($10^{-8} \Omega \cdot \text{m}$),^{202,203} it is believed that the deposited IrO_x on the titanium support provides the electronic conductivity within the CL. In principle, if a core-shell structure is achieved, there will be complete conductivity and IrO_x utilization. In practice, however, complete films of IrO_x on TiO₂ are not simple to prepare. Instead, electronically isolated islands of IrO_x on TiO₂ often result. Thus, to achieve high electronic conductivity within the CL, interconnected networks of IrO_x long enough to reach catalyst particles not in direct PTL contact would be required. This type of network would reduce the probability of electrically disconnected catalyst particles and thereby maximize catalyst utilization.

4.1.2.3 Electronically Conductive Networks

Two strategies have been employed to help enable electronically conductive networks: optimizing CL thickness and developing core-shell-supported catalyst structures.

CL thickness is a metric correlated with catalyst utilization, as a CL that is too thin will have less interconnected iridium particles and thus a lower probability of forming a complete in-plane (parallel to the membrane) electron conduction network throughout the layer. Hence, it is necessary to maintain a certain CL thickness to avoid such issues. Through cross-sectional SEM, the thickness of the CL is shown to be linearly proportional to iridium loading.²⁰⁴ Through experiments using Umicore Elyst Ir75 (IrO₂/TiO₂) with various Ir loadings between $1\text{--}2 \text{ mg}_{\text{Ir}} \cdot \text{cm}^{-2}$, Bernt et al.¹⁹³ proposed an optimal anode CL thickness of $4\text{--}8 \mu\text{m}$. The lower bound ($4 \mu\text{m}$) ensures adequate in-plane electronic conductivity, while the upper bound ($8 \mu\text{m}$) limits mass transport overpotentials, as thicker layers suffer from water transport issues. In a later study,¹⁹⁴ Bernt et al. calculated that a low packing density Heraeus P2X would require a loading of $0.06 \text{ mg}_{\text{Ir}} \cdot \text{cm}^{-2}$ to form a $4\text{--}8 \mu\text{m}$ thick CL, as opposed to the $0.56 \text{ mg}_{\text{Ir}} \cdot \text{cm}^{-2}$ required for Umicore TiO₂. While the logic of using larger support particles to enhance CL thickness (without increasing Ir loading) is sound, increased CL thickness does not necessarily equal increased CL conductivity if percolating catalyst networks are not present.

Several studies have sought core-shell structures where the percolation of IrO_x coatings would provide the needed electronic conductivity at low loadings.^{197,205,206} De Angelis et al.²⁰⁷ demonstrated connected films of IrO₂ supported on TiO₂ in a CL via X-ray ptychographic tomography (a non-destructive 3D imaging technique), although the loading was very high— $3 \text{ mg}_{\text{Ir}} \cdot \text{cm}^{-2}$. Given the well-connected pathways for IrO₂, the authors concluded that for “any CL where the percolation of the IrO₂ is ensured, any high-surface area support material can be employed, even if fully electrically insulating.” The concept has been extended to PTE configurations as well, with one study featuring IrO₂/TiO₂ core-shell catalysts ($0.4 \text{ mg}_{\text{Ir}} \cdot \text{cm}^{-2}$) that performed better than the Umicore IrO₂/TiO₂ catalyst ($0.5 \text{ mg}_{\text{Ir}} \cdot \text{cm}^{-2}$).²⁰⁸ While increased electronic conductivity is beneficial, it is possible that CL conductivity is not as critical for PTEs as for CCMs, given that the catalyst in PTEs is directly deposited on PTL particles, eliminating concerns of unutilized catalyst within PTL pores.

In summary, using TiO₂ as a support is not a guarantee of increased catalyst utilization as the insulating nature of TiO₂ can render iridium oxide particles electrically isolated and therefore unutilized, unless there is a robust network of connected iridium oxide domains. That said, there have been demonstrations of promising performance at loadings below $0.4 \text{ mg}_{\text{Ir}} \cdot \text{cm}^{-2}$, even after thousands of hours of testing.^{209,210} Further investigations in creating percolating iridium oxide pathways under low loadings

are warranted. Lastly, alternative materials and doped TiO_2 are showing promise and should be examined for long-term durability, as finding a conductive support (that is stable) would eliminate this percolation challenge.

4.1.3 Durability

While researchers seek to lower catalyst loadings to meet cost targets, care must be taken to ensure that durability is not compromised during the process. However, as reviewed by Wang et al.,¹⁶³ only six MEA studies have examined iridium durability at loadings $\leq 0.4 \text{ mg}_{\text{Ir}} \text{ cm}^{-2}$. Hence, mechanisms of degradation and parameters that exacerbate degradation still need to be identified for low-loading systems.

To understand iridium degradation mechanisms, Yu et al.^{173,176} conducted a microscopic analysis of an ultra-low loading CCM ($0.08 \text{ mg}_{\text{Ir}} \cdot \text{cm}^{-2}$) under industrial conditions ($1.8 \text{ A} \cdot \text{cm}^{-2}$ constant load, ~ 30 bar pressure, 86 cm^2 active area). They found that 70% of the initial iridium was lost after 4500 h of operation. Numerous other studies have also found significant iridium dissolution over time, with the membrane, cathode CL, and water streams (anode and cathode) being the primary sinks for subsequent redeposition.^{137,211–214}

Parameters shown to be highly degrading to the anode catalyst are OCV operation, dynamic operation, and low iridium loadings. Weiß et al.²¹⁵ investigated the impact of a 1.3 V hold versus an OCV hold—where voltage decreases to 0V—in a cycling profile. Two protocols were used (**Figure 9a**): An OCV test (3 A/cm², 0.1 A/cm², and OCV steps) and a reference test (3 A/cm², 0.1 A/cm², and 1.3V steps). The former resulted in more degradation due to the OCV conditions reducing crystalline IrO_2 (the starting material) to metallic Ir and then oxidizing it to amorphous IrO_x (which is less stable). Overall, they found a very high degradation rate of $\sim 300 \mu\text{V/h}$ for this test and, surprisingly, a negative degradation rate (performance increased over time) for the reference test (both used a high anode loading of $1.75 \text{ mg}_{\text{Ir}}/\text{cm}^2$). Incorporating the OCV hold instead of the 1.3 V hold led to faster iridium dissolution and increased HFR (due to the IrO_2 transforming to a less-conductive hydrous iridium oxide).

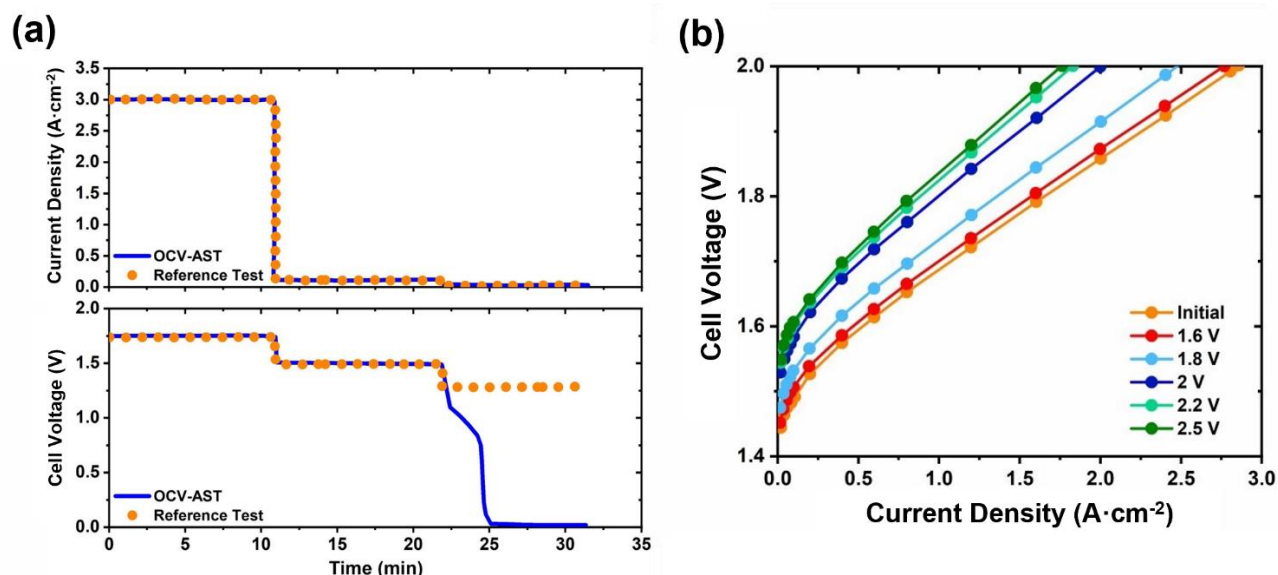


Figure 9. (a) Current density and voltage profiles (of one cycle) for the OCV test and reference test used to examine the impact of OCV holds on cell performance. Adapted from ref.¹³⁷ Copyright 2019 Journal of The Electrochemical Society under Creative Commons Attribution Non-Commercial No Derivatives 4.0 License (<http://creativecommons.org/licenses/by-nc-nd/4.0/>). (b) Polarization curves after 525 hours of operation with various square wave cycling protocols, from 1.45V to different upper potentials ranging from 1.6-2.5 V. Initial (i.e., beginning of life) polarization curve is shown in orange as a reference. Adapted from ref.²¹² Copyright 2019 Journal of The Electrochemical Society under Creative Commons Attribution 4.0 License (<http://creativecommons.org/licenses/by/4.0/>).

Regarding dynamic operation and loadings, Alia et al.²¹⁶ examined durability at low loadings (0.1-0.4 mg_{Ir}/cm²) and reported that potential cycling resulted in greater degradation than potential holds. Furthermore, square wave cycling was more damaging than triangle wave cycling, suggesting that sharper transitions accelerate degradation. They also found that greater cycling frequencies and higher upper potentials (**Figure 9b**) accelerate degradation. Moreover, the impact of catalyst loading on degradation was also investigated. While they observed minimal degradation at 0.4 mg_{Ir}·cm⁻², meaningful degradation was found at 0.1 mg_{Ir}·cm⁻². The authors hypothesized that degradation is not inherently loading-dependent, but rather, the extra thickness of the CL at higher loadings acts as a buffer that delays the onset of degradation. A 1000 h low-loading MEA stability test by Rozain et al.²⁰⁹ backs up this hypothesis, as they found that two CLs with similar thicknesses but a different catalyst and loading (IrO₂ at 0.1 mg_{Ir} cm⁻² and pure IrO₂ at 0.32 mg_{Ir} cm⁻²) had comparable durability.

Standardized protocols to characterize degradation will need to be developed to test durability more quickly (i.e., without having to operate cells over thousands of hours). Furthermore, standardization will allow the community to objectively compare MEAs with different loadings and morphologies. For instance, different fabrication methods (e.g., blade coated vs. spray coated) result in distinct CL morphologies and the nature of their degradation under low catalyst loadings can be significantly more sensitive.¹⁶³ Thus, universal protocols are a prerequisite for efficient research in developing next-generation MEAs. Recommendations for such protocols will be discussed in **Section 5.1**.

4.1.4 Ionomer Binders

As mentioned previously, CLs typically consist of catalyst particles dispersed in an ionomer network. The whole layer must expose the surface of catalyst particles for reactions, maintain charge conducting pathways, enable reactant transport to these catalyst sites, and remove reaction products—all of which are impacted by the ionomer. Please note that since ionomers are used for both the membrane and CL, “ionomer binders” will be used to refer CL ionomers only. In commercial MEAs, the ionomer binder has an identical/similar chemical composition to that of membrane ionomers. Some ionomer properties (such as ionic conductivity) are beneficially optimized in the same direction for membrane and binders. Other properties that are critical to membrane applications prove detrimental to binder applications, including high electrical resistance and low gas permeability. As there does not exist a single material appropriate for all properties, a similar (often identical) electrode ionomer is employed to enable ionic conduction within the catalyst layer, which has a strong impact on the overall optimization of catalyst layer ionic conductivity, electronic conductivity, and mass transport.

The GW-scale deployment of electrodes in PEMWE can leverage decades of fundamental understandings of catalyst/ionomer integration from PEMFC research. For example, Raistrick demonstrated that by replacing PTFE with an ionomeric binder, platinum utilization was improved to the point where loadings could be reduced by an order of magnitude (leading to their ubiquitous inclusion in PEM devices).²¹⁷ Baker et al.²¹⁸ and Greszler et al.²¹⁹ identified an unknown transport resistance in a fuel cell cathode associated with the ionomer, which was later demonstrated to arise from a local oxygen transport resistance due to ionomer orientation at the platinum/ionomer interface.^{220,221} The findings from these studies suggest the importance of considering a holistic set of properties of binder candidates and how they impact transport properties in PEMWE CLs, especially as iridium loadings are driven lower.

While much can be learned from PEMFC research efforts, novel electrolyzer-specific configurations are also being explored that have the potential to influence ionomer selection and where it is deployed. Where most electrolyzers feed water to the anode directly, other systems rely on back-diffusion from the liquid-fed cathode²²² or water vapor anode feeds,²²³ which could influence the optimal electrode composition at the anode in unexpected ways. For example, Peng et al.²²⁴ demonstrated a promising MEA strategy whereby a low loading of iridium oxide was deposited directly onto the PTL layer, avoiding the need for ionomer in the anode entirely. The role of ionomer in scalable, durable, and high performance electrodes in PEMWE systems will require further investigation to enable deployment at scale.

4.1.5 MEA Fabrication

Finally, large-scale production methods for low loading CLs requires more development. The majority of the low loading MEAs tested in the literature uses the ultrasonic spray coating technique, which requires a catalyst ink with relatively low solids ratios (< 1 wt%).^{175,177,181,225–227} However, this technique is not suitable for large-scale production due to its low throughput.²²⁶ Blade coating techniques on the other hand (e.g., doctor blade coating, rod coating, and slot die coating), are more scalable but require inks with higher solids ratios (> 5 wt%),²²⁷ which tend to result in inhomogeneous catalyst distribution when lower loadings are targeted. Thus, more research is required to enable large-scale fabrication of low loading MEAs, likely via blade coating techniques. For example, understanding the key parameters of catalyst inks that will allow for uniform CLs, and minimizing catalyst loss from ink intrusion into PTL pores during PTE fabrication. More examination in topics such as these will be essential in curtailing PGM usage.

4.2 Cathode Catalyst Layer

Platinum black supported on carbon (Pt/C) is the standard cathode catalyst for PEMWEs. Studies on Pt-free catalysts have been reviewed^{228,229} but remain an active research area. Given the sparsity of durability studies with such catalysts, this section will be limited to Pt-based catalysts.

Although studies utilizing Pt-based cathode catalysts typically assume negligible cathode contributions to cell overpotential, the true contribution may not be obvious, especially when considering open circuit conditions (relevant during startup and shutdown conditions). On the one hand, one model estimated cathode overpotentials (for a single cell) of less than one mV at 3 A cm^{-2} ²³⁰ and one experimental study found that performance was insignificantly affected by Pt loading reductions from $0.30 \text{ mg}_{\text{Pt}} \text{ cm}^{-2}$ to $0.025 \text{ mg}_{\text{Pt}} \text{ cm}^{-2}$.²⁰⁴ However, a modeling study of a 12-cell, 5.6 kW stack found that cathode overpotentials can reach up to 63% of the anode overpotential when the cathode pressure is increased to 70 bar.²³¹

4.2.1 Overpotentials

Cathode overpotentials were not experimentally verified in (single-cell) PEMWEs until the introduction of in-situ reference electrodes, which was recently reviewed.²³² In one study with such an electrode (3 mg cm^{-2} Pt black for the cathode, 3 mg cm^{-2} $\text{Ir}_{0.7}\text{Ru}_{0.3}\text{O}_4$ for the anode), when the current was switched from 1 A cm^{-2} to open circuit conditions, the cathode potential evolved from -0.06 V to 1.05 V and the anode potential from 2.00 V to 1.25 V (vs RHE).²³³ While these measurements confirm that the anode overpotential was indeed many times larger than that of the cathode, it also showed that the cathode experienced a more significant change in half-cell potentials during the transition from operation to open circuit conditions, which was attributed to Pt oxidation due to crossover oxygen (from the anode). A later study tracked the evolution of the cathode overpotential alongside in-situ Pt dissolution measurements via a hanging mercury drop electrode.²³⁴ The resulting heat map of Pt dissolution—as a function of load duration (at 1 A cm^{-2}) and open circuit potential duration—found the most degrading condition to be cycling between 0.01 h at 1 A cm^{-2} and 7.9 h at open circuit. Notwithstanding, a 1000 h study utilizing a five-cell stack found that a constant 2 A cm^{-2} hold resulted in larger cell potential increases than alternating between open circuit and 2 A cm^{-2} (for both 10 minutes and 6 hours at each current).²³⁵ These two studies are not necessarily in disagreement; even if Pt dissolution—and perhaps cathode degradation—is highest when alternating between operation and open circuit, the overall cell degradation was still more pronounced during operation, agreeing with the consensus that the anode defines the majority of degradation.

The only known AST study utilizing an in-situ reference electrode²³⁶ examined the evolution of half-cell potentials over 1000 h while employing a dynamic load degradation test (defined by the *EU Harmonized Protocols for Testing of Low Temperature Water Electrolysers*). While the anode experienced a substantial increase in half-cell potential during the first week and the cathode did not, the rates of half-cell potential increases were similar for both in the subsequent weeks. However, the authors noted that half-cell potential measurements were sensitive to even slight movements of the reference electrode, with many results potentially explained by either a physical change or a disturbance of the reference electrode. Unfortunately, these events are especially relevant when working with long-duration tests, where unanticipated shutdowns and adjustments are common.

4.2.2 Post-Mortem Analyses

Given the challenges of long-duration in-situ half-cell monitoring, post-mortem analysis becomes especially relevant to understanding cathode degradation, although results are sparse. A previously mentioned study (in **Section 4.1.3**) by Yu et al.¹⁷³ examined low-loading (0.3 mg cm^{-2} Pt, 0.08 mg cm^{-2} Ir) CCM degradation (4500 h of operation at 1.8 A cm^{-2}) with cross-sectional SEM coupled with EDS. Pt/Ir bimetallic particles were observed in the membrane (on the side closer to the cathode), and Ir particles were observed in and adjacent to the cathode CL. Although Ir degradation was the most pronounced—with 70% of the original Ir lost—Pt degradation was not negligible, with 10% of the original Pt lost (to the membrane), which was understood to be due to the transient reduction of Pt oxides at the beginning of the test. A similar study using cross-sectional SEM/EDS has also observed Pt and Ir particles in the membrane after 1000 h of dynamic loading testing.²³⁷ Lastly, Rakousky et al.²³⁸ used post-mortem TEM imaging and observed that cathode catalyst particle size increased from 3.5 nm to 7.8 nm after 1000 h of operation (at 2 A cm^{-2}). However, the authors concluded that it was unlikely that this particle growth significantly contributed to decreased cell performance, instead citing PTL corrosion and the anode activity reduction.

4.2.3 Outlook

In conclusion, the assumption that cathode overpotentials negligibly contribute to beginning-of-life single-cell performance is supported by experimental results, but it is unclear whether this holds for multi-cell stacks (especially at higher pressures). In-situ monitoring of half-cell potentials becomes especially challenging for extended studies due to unexpected adjustments/changes to the reference electrode. The studies presented suggest that cathode contributions to degradation are strongly masked by those of the anode for single-cell durability. Future single-cell durability studies could benefit from in-situ reference electrodes, although precautions must be made to ensure a stable reference electrode. Post-mortem studies can contribute substantially to understanding degradation processes, but there are still gaps in linking degradation to PEMWE performance loss; such a gap could be addressed with modeling studies. Additionally, future studies utilizing multi-cell stacks and higher pressures could benefit from more clearly elucidating the effects of cathode degradation on overall system durability. Such studies would benefit from the collaboration between academic institutions and industrial companies, with the former spearheading electrochemical/post-mortem studies while the latter provides the necessary equipment for such experiments.

4.3 Membrane

The type of membrane used in PEMWEs is a proton exchange membrane (PEM) and is responsible for conducting protons and preventing gas crossover. While the membrane neither contributes significantly to costs nor is a substantial factor for cost reductions through economies of scale (**Figure 7b**), membrane research is a primary lever for voltage efficiency and lifetime improvements.

State-of-the-art membranes are based on perfluorosulfonic acid (PFSA) chemistry. PFSA is a copolymer of tetrafluoroethylene (used in Teflon™) and a sulfonic acid-terminated perfluorinated monomer (**Figure 10a**). This combination of hydrophobic backbone and hydrophilic sidechain results in a phase-segregated structure in which cation-conducting water channels form and percolate through a solid matrix. Distinctions between PFSA membranes and hydrocarbon alternatives (**Figure 10b**) will be discussed in **Section 4.3.5**. For many decades, PFSA membranes have enabled PEMWEs to (1) operate at high current densities, (2) operate under dynamic electrical loads, (3) achieve high energy efficiency, (4) produce high purity hydrogen (up to 99.999%), and (5) achieve long lifetimes.²³⁹

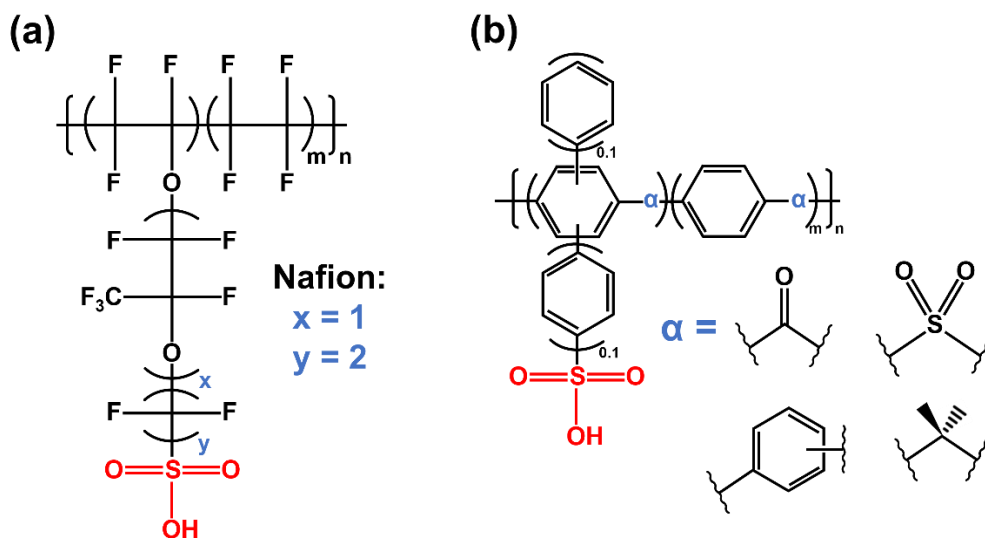


Figure 10. Prototypical structures of two proton exchange membrane (PEM) chemistries. (a) PFSA-based: a hydrophobic tetrafluoroethylene backbone with a hydrophilic side chain ending with a sulphonic group. (b) hydrocarbon-based: a network of rigid hydrophobic aromatic groups with sulphonic groups attached at some ends.

4.3.1 Targets

Membrane-related targets set forth by the U.S. DoE are shown in **Table 2**.²⁴⁰ State-of-the-art membranes (such as Nafion™ N115) have excellent durability (leading to high lifetimes) but elevated ohmic overpotentials due to their high resistance to proton transport. Reducing this inefficiency will likely involve some combination of a more conductive ionomer and a reduced membrane thickness. However, these changes can impose mechanical strength and gas crossover challenges, especially when operating at high differential pressures (≥ 30 bar).²⁴¹ Insights in improving performance in these areas will be summarized, along with perspectives on developing a composite membrane enabling 2026 (and beyond) DoE targets.

Table 2. Membrane-related targets for PEMWEs (80°C, 30 bar H₂ pressure).^{240,242}

Parameter	Units	Current Status (Nafion™ N115)	2026 Target
Lifetime	h	60,000	80,000
Degradation Rate	%/1000 h	25	13
Activity @ 3 A·cm ⁻²	V	2.0	1.8
PEM Thickness	μm	125	50
PEM Resistance	mΩ·cm ⁻²	100	30
H ₂ Crossover	% H ₂ in O ₂	2	0.5

4.3.2 Ohmic Losses

Improving voltage efficiency is largely incumbent on reducing membrane proton transport resistance. For a given operating condition (e.g., temperature), two factors can reduce this resistance: (1) improving ionomer conductivity and (2) minimizing membrane thickness.

The conductivity of a PFSA ionomer is a strong function of equivalent weight (EW), defined as the mass of polymer per mole of sulfonic acid (g/mol). In an ionomer review,²⁴³ conductivity as a function of EW for many ionomers was evaluated, and it was shown that many lower EW ionomers exist below that of Nafion™ N115 (1000 EW). However, a strong lower bound for EW exists for practical applications dictated by a given ionomer's water uptake/solubility. As water uptake increases from decreased EW, issues such

as low mechanical strength and high gas crossover can arise that may counteract the benefit of proton conductivity increases. Furthermore, the differences in ionomer conductivity from different EWs are minimized in the liquid-saturated PEMWE environment.²⁴⁴

Apart from a significant ionomeric breakthrough, much of the reductions in resistance will likely come from thinner constructions. As early as 2015, Lewinski et al.²⁴⁵ reported a 50 μm membrane achieving 1.77 V at 3 A cm^{-2} (80°C), which is $\sim$ 150-200 mV lower than for standard N115 membranes. Furthermore, Garbe et al.²⁴⁶ observed a $\sim$ 240 mV decrease at 3.0 A cm^{-2} when comparing a 183 μm and 50 μm membrane. However, making membranes thinner makes them more susceptible to mechanical deformation and gas crossover. Utilizing reinforcements can potentially alleviate these issues.

Membrane reinforcements have been used in more mature electrochemical devices (e.g., chlor-alkali and hydrogen fuel cells) for (1) improved mechanical durability (by giving resistance to crack formation and through-plane defect penetration), (2) lower expansion in water and other solvents, and (3) processability during manufacturing and cell assembly. For PEMWE, an early example of a reinforced membrane is the directionally stable membrane (DSMTM) developed at Giner, where holes were formed in a flat reinforcing material. While significant morphology optimization occurred, it is unclear whether an economical route to manufacturing DSMs at scale was ever realized.²⁴⁷ More recent examples include woven fabrics and cloths (based on fluorocarbon or hydrocarbon fibers)²⁴⁸ and non-woven meshes (such as expanded polytetrafluoroethylene (ePTFE) or electrospun materials).^{249,250} The ideal reinforcement is one that provides increased durability and reduced crossover while being able to be produced at a low cost and high volume.

4.3.3 Gas Crossover

Hydrogen gas crossover from the cathode to anode can lead to the formation of explosive mixtures as the lower explosive limit (LEL) of hydrogen in oxygen is just 4% (by volume). Thus, the greater crossover rate for thinner membranes is a major issue,²⁵¹ especially when hydrogen is compressed *in-situ*. This issue is exacerbated during periods of low electrical load (i.e., low current density), where oxygen content is minimal.⁴² Three main strategies can be employed to mitigate gas crossover: (1) improving membrane gas barrier properties, (2) implementing a recombination catalyst (RC) to react crossover hydrogen with oxygen (which forms water), and (3) utilizing BoP solutions such as pressurizing the anode stream or installing external air circulators to dilute crossover hydrogen. The membrane-based strategies—(1) and (2)—will be discussed herein; we refer the reader to a review by Stähler et al.²⁵² for a discussion of (3).

For gas barrier properties of a membrane, a common metric is the selectivity of protonic (H^+) versus molecular hydrogen (H_2) transport (gas crossover). Not only does a higher selectivity reduce crossover, but also increases the Faradaic efficiency of the cell (due to less cathodic hydrogen lost from crossover).²⁵³ Several studies have explored the nature of H^+/H_2 transport in ionomers.²⁵⁴ Two studies by Stolten et al.^{255,256} demonstrated that hydrogen crossover through the membrane is purely diffusive. Fumagalli et al.²⁵⁷ studied PFSA ionomeric d-spacing as a function of time and revealed that the size of water channels never reach steady state, suggesting that membrane crossover could increase over its lifetime. Future ionomer design could incorporate these learnings to enhance selectivity. One strategy to improve selectivity (besides using reinforcements) is to incorporate non-ionomeric fillers, such as graphene or graphene oxide, which are proposed to block gas crossover while enhancing proton transport.^{258,259}

In addition to improving gas barrier properties, strategies to mitigate existing crossover hydrogen will likely be necessary, as some level of crossover will always occur. Several strategies include a downstream hydrogen gas recombiner,²⁶⁰ a third electrode in the membrane,²⁶¹ and a recombination catalyst (RC). All three strategies combine crossover H_2 and O_2 to form water, but RCs are the most commonly studied. They can be implemented in several locations, including as a coating on the PTL⁴² or anode CL,²⁵² or within the anode CL²⁶² or membrane.²⁶³ Furthermore, RCs can expand the operating current range in which PEMWEs can safely operate.²⁶⁴

RCs within the membrane have been used for decades (first to increase humidity in fuel cell membranes^{38,265,266}) and have several advantages, including 1) isolating the RC from high anodic potentials and 2) mitigating both hydrogen and oxygen crossover (which minimizes peroxide formation at the

CLs).^{252,267} RCs are typically PGM-based and can be implemented in membranes via (1) reduction of PGM salts to form RC nanoparticles *in situ*²⁶⁸ or (2) direct incorporation of pre-formed RC particles.²⁷ The optimization of the RC location (within the membrane) and loading are active areas of study. For example, Gubler et al.²⁶⁹ studied three RC distributions in the membrane and found the most effective crossover mitigation occurred when the catalysts were uniformly distributed. While research on RC optimization is progressing, insights into the long-term effectiveness of RCs over system lifetimes are lacking. Furthermore, the cost and complexity (i.e., the number of unit operations) for manufacturing RC-incorporated membranes must be minimized for facile uptake in future PEMWE systems.

4.3.4 Durability

Membrane durability should be one of the most prioritized research efforts, especially as designs tend towards thinner membranes for efficiency improvements. As early as 1988, PFSA membranes have enabled PEMWE durability above 10,000 hours.²⁷⁰ In 2006, a paper reported lifetimes of PFSA-based devices above 50,000 hours at moderate temperatures (~50-60°C).²⁷¹ This paper illustrates that PFSA membranes have long enabled devices to be incredibly durable; devices that were manufactured almost two decades ago have lifetimes that nearly reach 2026 targets. However, the significant re-optimization of membrane construction necessary to meet performance targets (i.e., incorporating reinforcements, RCs, more conductive ionomers, and thinner membranes) will represent a significant challenge to achieving future durability targets.

Fortunately, there have been several recent reviews covering membrane durability in PEMWEs.^{48,272–275} A high-level summary of their findings and perspectives on the most promising future research efforts will be provided here. Membrane failure modes can be mechanical or chemical/electrochemical. Membrane creep is a mechanical mode associated with membrane swelling and PTL/gasket integration (for a visual of how membrane and PTL interact, refer to **Figure 8**). Other mechanical failure modes—such as pinhole formation and membrane dry-out—may arise from stack-level defects (e.g., inconsistent heating or water distribution; poor stack assembly). Chemical/electrochemical failure modes can also exist, including cation contamination and radical-induced degradation. Furthermore, multiple modes can be present simultaneously and could combine to accelerate degradation.²⁷⁶ Lastly, while not a failure mode itself, higher temperatures (which are used to reduce kinetic overpotentials and transport resistances) can accelerate the rate of each of these modes.^{271,276–278}

4.3.4.1 Membrane Creep

Membrane creep can occur in both active (participating in electrochemistry) and inactive (gasketed) areas of the cell, along with the interface between the two. Creep failures have been identified as a limiting factor in cell lifetime for thin membranes. For example, a study from Nel Hydrogen²⁷⁹ reported ~500 hr lifetime for a thin (89 µm) membrane compared to ~4500 hours for a thick (178 µm) membrane (both membranes were non-reinforced), where the lifetime gap was ascribed to membrane creep. Reinforcements have been identified as a solution for creep, as the same study also reported that a 60 µm reinforced membrane exhibited similar durability to the thick non-reinforced membrane.²⁷⁹ However, there is a lack of mechanistic understanding of this failure mode as well as methods to evaluate its potential impact. The successful deployment of thin membranes will depend on understanding and quantifying membrane creep. Work tying fundamental mechanical properties to observed creep failures will be critical toward mitigating these issues. Since creep might occur in a tensile mode (i.e., penetration into PTLs) and compression mode (as a result of compression under gaskets), multiple testing protocols might be required to evaluate creep in different cell areas.^{178,280}

4.3.4.2 Cation Contamination

Cation contamination leads to reduced protonic conductivity and increased overpotentials. Even with a pristine proton-form membrane at the beginning of life, several sources of contamination are conceivable, including feed water impurities and dissolution of metal cell components (e.g., PTLs and BPPs).²⁸¹ In principle, a small amount of infrequent cation contamination could be reversible, as the *in-situ* generation

of protons and excess circulation of liquid water provides a pathway for metal salts to exit the cell.²⁸² However, flowing a preponderance of metal ions into the ionomer material could render the stack highly inefficient or inoperable (which is possible, for example, through an acute upset of water quality). Theoretically, the ionomer's native protonic form could be refreshed by circulating an external acid solution.²⁸³ The practicality of this approach at GW scales is unclear, and it could potentially extract beneficial additives such as RCs or radical scavengers (discussed in the next paragraph). The most cost-effective strategy to remediate ion contamination is to prevent it in the first place, as is often employed in water recirculation loops in the form of a combination of reverse osmosis and ion exchange resins.²⁷⁵

4.3.4.3 Radical-Induced Degradation

The crossover of H_2 into the anode or O_2 into the cathode can lead to the formation of H_2O_2 , which acts as the basis for radical species generation.²⁸⁴ Certain metals—such as Fe, Ti, Cr, Ni, Cu, Pt, and Ir—can catalyze H_2O_2 into harmful radicals such as $H\bullet$, $HO\bullet$, and $HOO\bullet$.^{37, 49 48,284} The reaction of these species have been documented as a central degradation mechanism in PEMWEs (and PEMFCs),²⁸⁵ but significant effort is still required to understand the fundamental nature of such degradation. Possible reactions of these radicals with PFSA ionomers are depicted in **Figure 11**. It should also be noted that changing the chemistry of the PFSA ionomer (such as adding bis-sulfonimide groups in the side chain) can potentially cause low stability against radical attacks.²⁸⁶

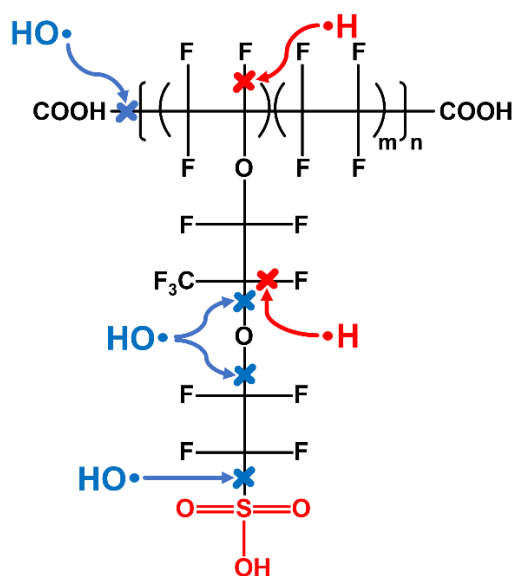


Figure 11: Possible radical-induced degradation mechanisms for PFSA ionomers. Sources: $HO\bullet$ attack on $-COOH$,^{287,288} $HO\bullet$ attack on ether linkages,²⁸⁹ $HO\bullet$ attack on $-SO_3H$,^{289,290} and $H\bullet$ attack on C-F bonds.^{291,292}

To quantify radical-induced degradation, measurements of fluoride anions liberated as a consequence of these reactions are commonly done, known as fluoride release rate (FRR) or fluoride emission rate (FER). FRR has been proposed as an overall metric for quantifying membrane thinning, as

the two have been reported to be directly correlated.²⁷⁷ Measuring FRR is typically done by ion chromatography or ion-selective electrodes. In one example, Fouda-Onana et al.²⁷⁷ measured FRRs at the anode and cathode effluent water of a PEMWE cell operating at 60°C and 80°C. Substantially higher FRRs were observed at the higher temperature as well as in the cathode effluent water, which was attributed to crossover oxygen reacting to form H₂O₂ in the presence of the platinum catalyst. Simpler ways to measure membrane degradation may exist, as Marocco et al.²⁹³ found a linear relationship between FRR and water conductivity by using a simple conductivity meter. This approach may be a way to monitor stack degradation at scale, though it would require removing or quantifying other contributors to water conductivity.

Over time, radical-induced degradation could become significant enough to thin the membrane significantly, exacerbating gas crossover and potentially causing electrical shorting. However, a wide range of outcomes are reported for the magnitude of radical-induced membrane degradation. For example, Grigoriev et al.²¹³ reported that membrane thickness was reduced by 75% over 5500 hr. Another study²⁹⁴ loosely accredited the observed 35 $\mu\text{V/hr}$ degradation rate to ionomer degradation (attributed to increased HFR). These results contrast with other studies where membrane degradation is not observed or is minor.^{270,271} To reduce the possibility of contradictory data, researchers must minimize the impact of degradation from other cell components (e.g., the PTL or aCL). It should also be noted that the degradation impact of different operation protocols (e.g., low current, high current, load-following) is an area of active study and may contribute to discrepancies in rates of radical-induced degradation.

Mitigating radical-induced degradation in PEMFCs has been achieved through (1) fluorinating the end groups of ionomers (currently standard in PEMWE membranes such as Nafion™ N115) and (2) including radical scavengers (cerium/manganese-based) in the MEA. Although radical scavengers have been reported to have benefits in PEMWEs,²⁹⁵ they have also been attributed to catalyst poisoning and increased mass transport overpotentials.²⁹⁶ Recombination catalysts may limit chemical degradation by lowering crossover gas concentration at the CLs (reducing potential routes to H₂O₂ formation), but their use has been reported to increase FRR in one study.²⁹⁵ Either way, improving the fundamental understanding of chemical degradation and the tradeoff of implementing various mitigation approaches will be necessary to ensure successful GW-scale deployment.

4.3.5 Hydrocarbons

Hydrocarbon-based membranes have long been sought as an alternative to PFSA ionomers owing to their potential for lower gas permeability, fluorine-free chemistry, and lower cost.²⁹⁷ Most hydrocarbon ionomers consist of a chain of aromatic units linked by various functional groups (**Figure 10b**). Although hydrocarbon membranes have promising attributes that can improve PEMWE performance, challenges in manufacturing and durability must be addressed to meet system targets. This section will outline the main obstacles for hydrocarbon membrane deployment and strategies to overcome them.

4.3.5.1 Water Uptake

Hydrocarbon ionomers tend to possess lower proton conductivity than PFSA ionomers given the same water volume fraction.^{298,299} One reason for this property is a result of their conductive groups (sulfonates) being connected to a rigid aromatic backbone, which resists phase segregation into conductive percolating pathways. Decreased dielectric screening and a lower degree of dissociation of the sulfonic groups in hydrocarbon ionomers also limits the proton mobility. As a result, greater water uptake is needed to achieve comparable ionic conductivity (e.g., a prominent hydrocarbon ionomer—sPPS—has a water volume fraction of 70%, while Nafion™ N115 has 40%³⁰⁰). However, this increased uptake makes them more prone to mechanical damage.

Several mechanical issues from high water uptake arise from the membrane's reduced dimensional stability, which increases delamination and embeds stress on the membrane structure.³⁰¹ Additionally, since there are spatial restrictions in the PEMWE cell that dictate the allowable extent of expansion, residual stress can build up, particularly along edges and under differential pressure (especially for thin membranes).^{302,303} Furthermore, higher water uptake can decrease mechanical toughness and reduce the gas permeation advantage of hydrocarbons, as the gas permeability of water is much higher than that of

the un-hydrated polymer.³⁰⁴ Lastly, mechanical degradation can feedback into chemical degradation through factors such as 1) increased ionomer surface area for reactions, 2) thermal concentration in deformed areas, and 3) exacerbation of radical generation through increased gas permeation and embedded stress.^{305–310}

Fortunately, strategies to alleviate water uptake issues have been found. The first strategy is to utilize membrane chemistries that require less water uptake to achieve a sufficient level of conductivity. For example, block copolymer-based hydrocarbon ionomers can efficiently aggregate conductive sites (at some sacrifice of permeability) to lower the required water uptake while maintaining conductivity.²⁶³ Secondly, reinforcements (that are strong and acid-resistant) can be added to reduce swelling as well as improve mechanical durability. Analogously to what has been applied for PFSA membranes, porous PTFE-based reinforcements have enhanced dimensional stability—and thus mechanical durability—for hydrocarbon systems.^{311,312}

4.3.5.2 Radical-Induced Degradation

Though hydrocarbon membranes possess lower gas permeability than PFSA counterparts, the significantly higher level of chemical degradation—due to the rapid hydroxyl attack of hydrocarbons—may offset this benefit.^{300,304,313,314} While one can slow this degradation with radical scavengers,³¹⁵ the attack is excessively fast to the extent that chemical degradation is challenging to prevent thoroughly without excessively compromising ionic conductivity.³¹⁶ Thus, to successfully benefit from the advantages of hydrocarbons, researchers must also weigh their disadvantages. Given that radical attack might be too rapid to be effectively prevented using scavengers, novel strategies to bolster chemical resilience should be explored, such as 1) performing polymer repair, 2) incorporating interfacial PFSA layers (e.g., in the membrane-CL interface), and 3) localizing radical scavenging at points of generation (i.e., catalyst sites).³¹⁷

Overall, when pursuing efforts to mitigate outstanding challenges of hydrocarbon membranes, care should be taken to ensure that strategies are mutually compatible (i.e., not causing harm to other system components). Additionally, the mitigation methods employed must be durable over the lifetime of the PEMWE, which can be challenging to test.

4.3.5.3 Hydrocarbon Binders

Although binders pertain to the CL and not the membrane, they will be discussed here as this section covers hydrocarbon ionomers as a whole. As discussed previously, hydrocarbon ionomers may offer some technical upside in membranes, but there is not an obvious benefit in binders. Thus, the development path for hydrocarbon binders will likely require significant effort to meet performance targets.

Hydrocarbon binders suffer from a multitude of issues, including 1) kinetic inhibition, 2) sluggish mass transport properties, 3) poor mechanical and chemical stability, and 4) CL manufacturing difficulties. For kinetic inhibition, while both PFSA and hydrocarbon ionomers contain sulfonate groups that can adsorb—and subsequently block—catalyst sites, the latter has been shown to be more inhibiting.³¹⁸ This difference is attributed to the greater presence of sulfonates and the poisoning action of phenyl groups³¹⁹ (since hydrocarbon ionomers require more to achieve comparable PFSA conductivities) and to phenyl group adsorption on catalyst sites.^{320–322} Regarding mass transport, binders must rapidly facilitate gaseous transport, as lingering gases could inhibit the HER and OER.^{323,324} While the lower gas permeability of hydrocarbons benefits membranes, it is detrimental for binders and leads to increased gas transport (i.e., mass transport) resistances.^{325–327} Poor mechanical stability is tied to high water uptake, which reduces dimensional stability and toughness, while poor chemical stability is due higher levels of radical-induced degradation.³²⁸ Furthermore, delamination of the CL is more commonly reported for hydrocarbon binders, causing increased contact resistance and decreased catalyst utilization.^{329 304,330,331} Lastly, CL manufacturing is also an issue. While PFSA binders readily form percolating networks, hydrocarbon binders have been linked to blocked pores (for mass transport)^{332,333} and electronically isolated networks, leading to higher mass transport losses and lower catalyst utilization.^{333–336}

Although many issues hinder hydrocarbon-based binders, many research levers exist to improve their efficacy: 1) polymer structure/property optimization, 2) solvent system optimization, 3) interfacial

engineering, and 4) alternative designs. Researchers can consider how structural properties—such as pore structure, thickness, and tortuosity—can be tuned to facilitate mass transport. Chemical properties, such as conductivity and water uptake, can also be adjusted to enable greater mechanical strength. Furthermore, varying structural motifs (e.g., phenyl, ketone, etc.) can lead to less catalyst poisoning, reducing kinetic overpotentials. Optimizing solvent system parameters (i.e., solvent type, solid ratio, I/C ratio) to ensure high dispersity is critical to fabricating a well-connected microstructure. Tuning molecular weight of the binder could also help, as this property has been linked to aggregate formation and connectedness.³³⁷ Penultimately, interfacial engineering can be promising in combatting delamination and increasing interfacial contact.^{314,317,338} Lastly, alternative designs can be utilized, such as the ionomer-free PTE proposed by Lee et al.,²²⁴ or simply using PFSA binders with hydrocarbon membranes.³¹⁷

4.4 Porous Transport Layer

PTLs in PEMWEs are responsible for 1) facilitating $\text{H}_2\text{O}/\text{O}_2$ transport, 2) providing electrical conductivity between the CL and BP, 3) removing heat from the CL, and 4) mechanically supporting the membrane during differential pressure operation. For visualization, we encourage the reader to refer to **Figure 1** and **Figure 8**. PTLs significantly impact voltage efficiency since electrical and reactant/product transport through the PTL influences ohmic and mass transport overpotentials, respectively. Additionally, the PTL-CL interface has been found to influence catalyst utilization and, thus, kinetic overpotentials.^{339,340} All these factors underline the criticality of the PTL for performance. One note is that PTLs primarily facilitate through-plane (i.e., perpendicular to the membrane) transport, as opposed to BP flow fields, where in-plane transport dominates. Based on this terminology, a distinction between a standalone PTL and an integrated PTL+BPP design (typically used to reduce part count) can be drawn, as only the area where bulk through-plane transport occurs is considered the PTL for the integrated design.

PTLs are often compared to carbon GDLs in PEMFCs regarding functionality. However, the oxidizing conditions (i.e., >1.23 V) of the PEMWE anode do not allow carbon to be used due to it undergoing corrosion at potentials greater than 0.2 V.³⁴¹ State-of-the-art PTLs are made of titanium due to its stability under such conditions. Still, challenges of surface passivation (via oxidation) persist, which results in decreased electrical conductivity. The porous property of PTLs is due to the titanium being produced as felts, meshes, foams, or sintered powders.³⁴² More recently, the addition of a microporous layer (MPL)—a more porous/less tortuous layer at the surface of the PTL—is being developed to enable improved CL-PTL contact, which can enable lower catalyst loadings and thinner membranes. Although several PTL types have been studied, the final choice for stack integration will not only depend on performance but also on cost and scalability.

4.4.1 Cost

PTLs contribute $\sim 20\%$ of stack cost, or $\$47/\text{kW}$, when considered in a 1 MW system (**Figure 7**). However, through economies of scale for 1 GW/yr deployment, that cost can be essentially halved to $\sim \$25/\text{kW}$.³⁴³ Another significant cost lever is mitigating PGM usage for coatings. For passivation protection, PTLs are currently coated with platinum and iridium, which significantly adds to material costs. Given the limited understanding of degradation mechanisms, coatings are currently overengineered to ensure durability.^{344,345} Considering even a modest platinum coating of $0.1 \text{ mg}/\text{cm}^2$, the material alone can exceed $\$1.5/\text{kW}$ (assuming Pt cost is $\$50/\text{g}$).

Aside from initial material costs, there are significant expenses for managing and refining materials. Each instance of material handling can incur a substantial cost, which can be particularly daunting given the ambitious benchmarks required to meet the US DoE's target of $\$2/\text{kg H}_2$ by 2026. Fortunately, cheaper alternative base materials and non-PGM coatings have shown promise. For example, Stiber et al.³⁴⁶ demonstrated minimal degradation over 1000 h using a stainless steel PTL with a non-PGM coating (Nb/Ti).

4.4.2 Development

The various stages of PTL development are shown in **Figure 12**. *Step one* involves structure development. PTLs can be created through 1) thermal sintering, selective laser sintering, or electron beam melting of titanium powders, 2) heat treatment-induced diffusion bonding of titanium fibers, 3) weaving or expanding titanium meshes, and 4) tape casting or vacuum plasma spraying titanium particles.^{347–349} The most common method is powder or fiber sintering. Morphological properties—such as porosity, particle size, and thickness—are controlled by tuning process parameters (e.g., laser power, rastering rate, temperature) and starting material properties (e.g., particle size). Lastly, morphology can be quantified using techniques such as X-ray tomography³⁵⁰, mercury intrusion porosimetry³⁵¹, or optical profilometry.²²⁵ The influence of structure on performance is discussed in next section.

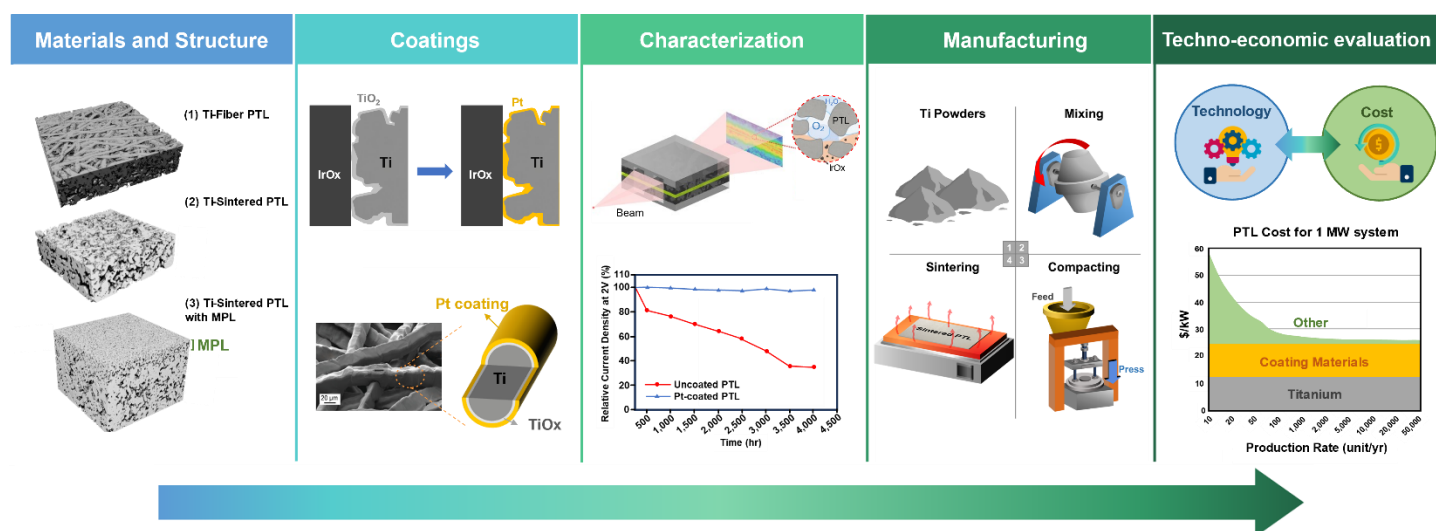


Figure 12. PTL development stages. PTL structure (1) adapted from ref.³⁵² PTL structures (2) and (3) adapted from ref.³⁵³ Copyright 2023 American Chemical Society. PTL coating figure adapted from ref.³⁵⁴ Copyright 2021 Journal of The Electrochemical Society under Creative Commons Attribution 4.0 License (<http://creativecommons.org/licenses/by/4.0/>). SEM micrograph and uncoated vs. coated PTL durability figure adapted from ref.³⁵⁵ Copyright 2021 Advanced Energy Materials under Creative Commons Attribution 4.0 License (<http://creativecommons.org/licenses/by/4.0/>). Interface characterization figure adapted with permission from ref.³⁵⁶ Copyright 2020 Journal of Power Sources. Tape caster image reprinted with permission from ref.³⁴⁸ Copyright 2019 Advanced Engineering Materials. Techno-economic data adapted from ref.¹⁴⁹ Copyright 2019 National Energy Renewable Laboratory (NREL).

Step two is the development of PTL surface coatings for corrosion resistance. State-of-the-art materials for coatings include precious metals such as platinum, gold, and iridium. Coatings will be discussed extensively in **Section 4.4.7**. Afterwards, characterization is done in *Step three* to benchmark activity and durability. The authors recommend examining how PTL-related properties (e.g., porosity/tortuosity, thickness, PTL-CL contact, membrane delamination into PTL, etc.) affect performance during this step.

Transitioning from lab-scale to industrial-scale, *step four* is manufacturing optimization for large-scale deployment. Although impressive morphological control can be achieved in lab scales, scaling up manufacturing (e.g., deploying roll-to-roll methods or creating batches >1000 cm²) can present quality control difficulties. Two methods are usually employed for large-scale manufacturing: 1) sintering—which involves combining, compacting, and sintering titanium powder, binders, and lubricants together, and 2) tape casting—which entails casting a titanium slurry into a thin layer, followed by drying and sintering. Tape casting provides precise control of sheet thickness and surface quality.³⁴⁸ For effective manufacturing, in-line quality control systems are necessary to guarantee morphology specifications and remove defective parts. Specifically, surface quality, presence of impurities, undesired edge warps or burrs, and inhomogeneous morphology need to be inspected. Failing to address these manufacturing-level defects

can result in stack-level failures that are much more expensive to address. Creating quality compliance standards and methods to identify defects are necessary research areas.

Lastly, *step five* is techno-economic evaluation. To maximize PTL market readiness, costs and manufacturing lead times should be minimized. Significant cost reductions can come from using non-titanium base materials and reducing PGM loadings for coatings. However, even if such improvements do not come to fruition, cost reductions can be achieved with higher production rates.³⁴³ For example, PTL cost can be almost halved from ~\$45/kW to ~\$23/kW by increasing production rates from 10 to 100 units/yr (assuming 1 MW units; refer to **Figure 7**).

4.4.3 Structure-Performance Relationships

Several studies have evaluated PTL structures and their effect on PEMWE performance. Apart from sintered powder and titanium fiber PTLs, researchers have studied meshes,³⁵⁷ expanded meshes,³⁵⁸ foams,³⁵⁹ as well as multi-layered combinations of these.³⁶⁰ Each PTL type can result in different interfacial contact behaviors with the CL, depending on the form factor of the surface. For example, the interface formed via sintered powder PTLs differs from that formed via sintered fiber PTLs, resulting in different performances even if their bulk properties (e.g., porosity and mean pore size) may be the same.

In a comprehensive review, Yuan et al.³⁴² analyzed the impact of PTL morphology on cell voltage and current densities (using sintered titanium powder PTLs). The results reveal a scattered relationship between mean pore diameter and voltage (**Figure 13a**). This scatter could be attributed to variations in test parameters among the different studies (e.g., temperature, pressure, and membrane thickness); nonetheless, a weak logarithmic correlation was observed, indicating that smaller pore sizes may decrease cell voltage. When examining porosity versus cell voltage (**Figure 13b**), higher porosity was loosely correlated with decreased voltage, but only at lower current densities of 0.2 and 1 A/cm². At 2 A/cm², no correlation was clear.

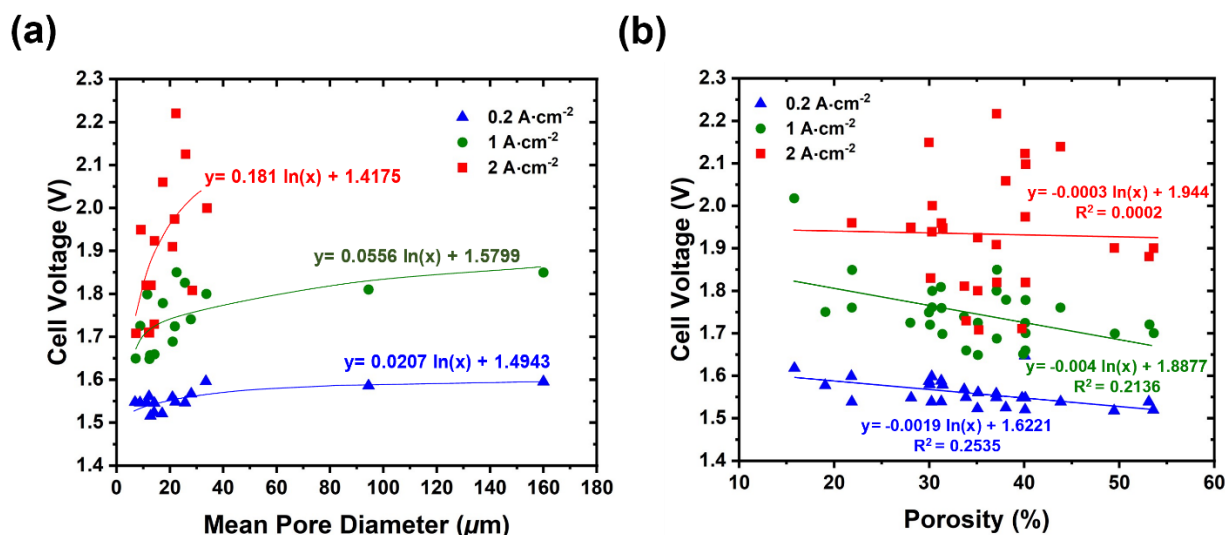


Figure 13. PTL structure-performance relationships. Mean pore diameter (a) and porosity (b) versus cell voltage at 0.2, 1, and 2 A/cm². PTL type was single-layer sintered titanium powder. Adapted with permission from ref.³⁴² Copyright 2022 Sustainable Energy & Fuels.

4.4.4 CL-PTL Interface

The CL-PTL interface is vital for PEMWE performance, especially under low loadings (as discussed in **Section 4.1.1**). The interface can be analyzed by calculating the interfacial contact resistance (ICR), which is influenced by contact pressure and deformation between the CL and PTL. For example, Yuan et al.³⁶¹ calculated ICR as a function of contact pressure and density of interfacial contact points. To directly estimate electrochemical performance, one can measure the triple phase contact area (TPCA), which quantifies the area common to the catalyst, ionomer, and PTL. Higher TPCAs result in higher catalyst utilization and lower kinetic overpotentials. Kulkarni et al.³³⁹ took this concept further and calculated ECSA with TPCA values obtained through X-ray computed tomography (CT). Using a fiber PTL and low iridium loadings (0.05-0.1 mg/cm²), Peng et al.¹⁷⁷ observed that very low TPCAs resulted in limited electron conductivity while very high TPCAs limited water accessibility to the CL (due to the catalyst being buried under the PTL fibers). Hence, for fiber PTLs with low iridium loadings, the lowest kinetic overpotential was observed at intermediate TPCAs between 35.4% and 29.4%, corresponding to a PTL porosity between 60%-62%. Higher catalyst utilization was also shown to reduce mass transport overpotentials, as higher utilization provides well-connected pathways between the CL and PTL for oxygen transport. Work by Schuler et al.¹⁷⁸ emphasizes this point by showing that mass transport limitations were more related to catalyst utilization than bulk mass transport.

Many modeling techniques have aimed to optimize the CL-PTL interface, given its importance in overall PEMWE cost and performance. The types of models used include one-phase flow, two-phase flow, image-based, and stochastic. A single-phase (water) 1D model was developed by Ma et al.³⁶² to simulate the effects of PTL pore size (at the interface) on catalyst utilization. A single-phase model sufficed since they focused on kinetically-dominated regions, where two-phase flow effects can be assumed negligible. They found that smaller PTL pores at the interface and better in-plane electrical conductivity in the CL improve catalyst utilization. A two-phase 3D model developed by Weber et al.³⁶³ investigated membrane hydration and catalyst utilization underneath PTL land and pores. The model showed 1) low catalyst utilization underneath PTL pores due to poor in-plane conductivity and 2) local dry-out of the membrane under PTL land with increased current density (**Figure 14**). Sepe et al.³⁶⁴ recently developed an image-based multiscale modeling method that uses X-ray CT images to resolve the PTL microstructure for use in two-phase flow simulations. Using these simulations, the model generates macroscale parameters—such as relative permeability and capillary pressure-saturation curves—to predict PEMWE performance. They found that the PTL with small pore sizes, a narrower pore size distribution, and the highest area of water-oxygen contact performed the best. They explained that smaller pores require less pressure for oxygen to enter, leading to lower mass transport losses. Lastly, Lee et al.³⁵⁷ used a stochastic pore network model to optimize reactant transport and interfacial contact. By tuning porosity and particle size, their results showed an optimized PTL with a porosity of 26.5% and a powder diameter of 25 nm under “normal” operation (exact operating conditions were not specified). Overall, much insight can be gained from the numerous CL-PTL interface models in the literature. However, future models need to include catalyst loading effects, as interfacial contact can suffer dramatically at low loadings.

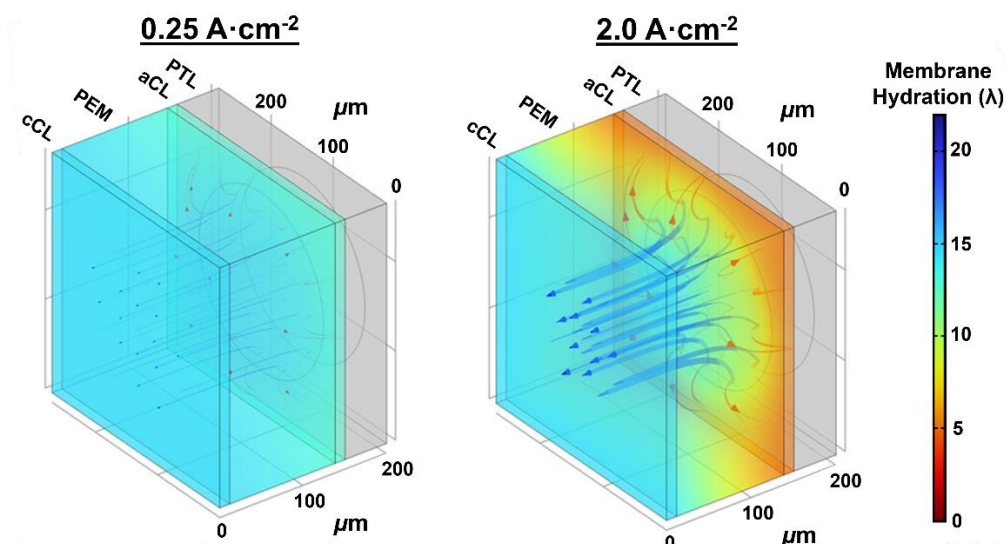


Figure 14. Contour plot showing membrane hydration underneath a single circular pore at two current densities. The streamlines represent contributions of water flux from electro-osmotic drag (blue) and diffusion (red). Adapted from ref.³⁶³ Copyright 2023 American Chemical Society.

Significant catalyst layer and membrane delamination can occur at the PTL-CL interface, resulting in electronically isolated catalyst islands that may not contribute to the OER (discussed in **Section 4.1.1**).¹⁵⁴ This effect is magnified under low loadings where fewer catalytic sites are available.^{178,180,367} Schuler et al.³⁶⁸ examined CL deformation post-cell operation and observed microcracks caused by stress imposed by the PTL (**Figure 15a**). The cracks formed below the fibers (highlighted in green) do not impede electronic conductivity since no PTL void space is present. However, the cracks next to the PTL void space (highlighted in red and yellow) do interfere with in-plane electronic conductivity, resulting in higher resistances. Furthermore, with differential pressure operation, CL/membrane creep into the PTL surface pores can become severe and propagate existing cracks in the CL. Over thousands of hours, creep can result in pinhole formation, especially when thin membranes are paired with PTLs with highly porous surfaces. A recent study by Ferner et al.³⁶⁹ employed nano X-ray CT and pFIB-SEM to investigate how CL structure changes over time. The study observed that 1) CL thickness decreased, and 2) CL regions compressed by PTL fibers (i.e., areas under and directly between PTL fibers) exhibited significantly different porosity and pore sizes compared to CL regions in the PTL pores (**Figure 15b** and **Figure 15c**). The compressed areas had a 20% decrease in porosity (~41% to ~51%) and a 50% decrease in maximum pore size (~600 to ~300nm).

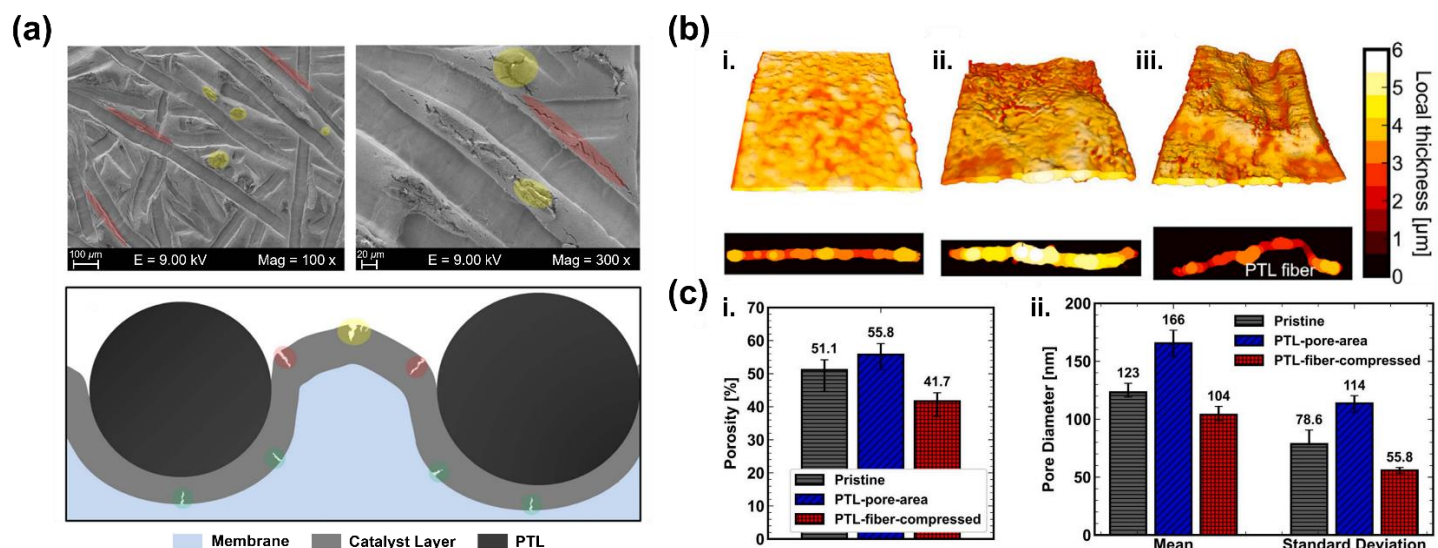


Figure 15. Membrane and catalyst layer deformation due to PTL compression. (a) Top: Post-operation SEM images of the anode CL surface. Bottom: schematic of CL deformation. Tension-induced microcracks (red) and compression fractures (yellow) are shown. Adapted from ref.³⁶⁸ Copyright 2019 Journal of The Electrochemical Society under Creative Commons Attribution Non-Commercial No Derivatives 4.0 License (<http://creativecommons.org/licenses/by-nc-nd/4.0/>). (b) 3D visualizations and 2D through-plane cross-sections showing thickness distributions for i. pristine spray-coated CL, ii. pristine blade-coated CL, and iii. tested blade-coated CL. Images obtained using X-ray nanotomography. (c) CL porosities (i.) and pore size distributions (ii.) in a pristine PTL, in PTL pores, or under PTL fibers. Adapted from ref.³⁶⁹ Copyright 2024 International Journal of Hydrogen Energy under Creative Commons Attribution 4.0 License (<http://creativecommons.org/licenses/by/4.0/>).

Optimizing CL-PTL interfacial properties will be essential for reaching high-performing low-iridium loading PEMWEs, perhaps even more so than bulk PTL properties. Lopata et al.³⁷⁰ revealed that interfacial properties—including pore diameter and titanium grain sizes—substantially impact catalyst utilization at low loadings and confirmed that interfacial pore properties were more influential than the bulk pore properties. Novel methods can be explored to improve interfacial properties of the PTL, such as laser ablation, pore patterning, and laser-induced structuring.^{225,371,372}

4.4.5 Mass Transport

Establishing efficient water transport and oxygen removal is important for high performance. Ambiguities have arisen in the definition of "mass transport" losses, with the term often used to encompass residual losses not associated with kinetic or ohmic overpotentials. However, we will exclusively use the term to refer to losses associated water and gas transport. Efforts to characterize mass transport in the PTL include operando X-ray imaging,^{373,374} neutron imaging,^{375,376} optical imaging,³⁷⁷ and numerical modeling,^{378–380} all of which have provided invaluable insights on maximizing performance.^{180,381,382}

The formation of oxygen bubbles can critically impede CL function and has been associated with mass transport losses.^{383,384} Such losses are more significant under low loadings and high current densities,^{175,177,225} where the increased amount of evolved oxygen gas could mask the already limited number of reaction sites. In their modeling study, Moore et al.³⁸³ found that mass transport losses can be caused by discrete transient events of catalyst site blockage by oxygen bubbles, even though gas saturation does not significantly affect liquid saturation across the CL. Bubble formation comprises three stages: nucleation on the CL surface, growth to a critical radius, and detachment. Lee et al.³⁸⁴ identified a "critical current density," where a sudden increase in gas saturation—and subsequently mass transport overpotential—occurs. They were able to increase the CCD by increasing the water flow rate, suggesting that there is a minimum flow needed for efficient oxygen bubble removal, as seen in **Figure 16**.³⁸⁵ Regarding heterogeneity, bubble accumulation can lead to current density and temperature maldistributions. Fortunately, optimizing the bubble transport properties of the PTL can mitigate the aforementioned detrimental effects.

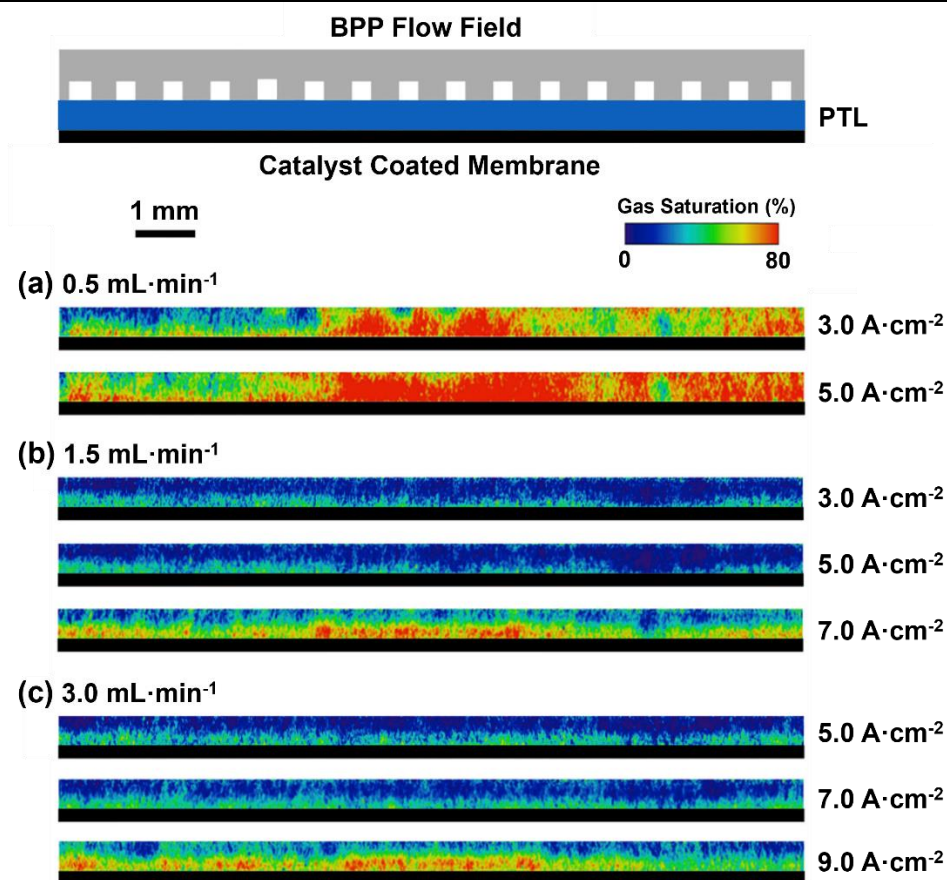


Figure 16. Neutron imaging of oxygen content in PTLs at different flow rates and current densities. A general schematic of the imaged region is at the top. Three oxygen profiles under various flow rates are shown in (a-c). The bottom of each image represents the CL-PTL interface while the top represents the PTL-BP interface. Adapted with permission from ref.³⁸⁵ Copyright 2020 Cell Reports Physical Science.

PTL gas transport can be enhanced by tuning wettability or optimizing PTL porosity and tortuosity.¹⁷⁷ Increased wettability has been shown to promote oxygen removal. Zhao et al.³⁸⁶ treated a PTL to increase its hydrophilicity and found increased activity compared to a control PTL. Furthermore, lower gas saturation was found in the treated PTL (through neutron imaging). Developing PTLs with higher porosity and lower tortuosity have shown reduced mass transport losses.¹⁷⁷ However, the balance of mass transport and interfacial contact needs to be carefully tuned, as studies have shown an inverse relation (at loadings $<0.1 \text{ mg}_{\text{Ir}}\cdot\text{cm}^{-2}$).³⁸⁷ Aside from developing new PTLs, modifying existing PTLs is another approach. Bazylak et al.³⁸⁸ demonstrated that creating perforations promotes faster gas removal and water intake. While having perforations present all the way through the PTL may negatively impact interfacial contact, Lee et al.²²⁵ showed that perforations that do not traverse all the way through can also improve mass transport without sacrificing interfacial contact. Efforts to reduce mass transport losses without compromising interfacial contact are required to satisfy performance targets, and the authors foreshadow that MPLs may be a solution.

4.4.6 Microporous Layer

To improve CL-PTL interfacial contact, using an MPL—a layer with smaller pore sizes than the bulk PTL—is proposed. Improved interfacial contact can lead to decreased mass transport and ohmic losses as well as increased catalyst utilization.

Numerous studies have examined the impact of MPLs on mass transport and ohmic losses. Using X-ray CT, Kulkarni et al.³⁵³ demonstrated that a thin ($\sim 20 \text{ }\mu\text{m}$) MPL on a PTL allowed for 20% increased

interfacial contact compared to a baseline PTL, improving mass transport properties. During the 1000 h stability test where the current was held at 1.8 A cm^{-2} , this MPL showed a $>100 \text{ mV}$ performance improvement and a lower degradation rate compared to the baseline PTL (at both catalyst loadings of $\sim 0.1 \text{ mg}_{\text{Ir}}/\text{cm}^2$ and $\sim 2 \text{ mg}_{\text{Ir}}/\text{cm}^2$). In a pioneering attempt to develop MPLs, Lettenmeier et al.³⁸⁹ vacuum plasma sprayed fine titanium powders (grain size $< 45 \mu\text{m}$) on sintered titanium powders. The resulting structure showed a clear bilayer, with the MPL and bulk PTL possessing pore sizes of 1.3 mm and 17 mm , respectively. The MPL resulted in marked ohmic improvement compared to the non-MPL, with a staggering HFR reduction of $20 \text{ m}\Omega \text{ cm}^2$. Following that, Kang et al.³⁹⁰ developed two MPL types, a microporous MPL using $\sim 5 \mu\text{m}$ particles and a nanoporous MPL using $30\text{--}50 \text{ nm}$ particles. Interestingly, they found that the microporous MPL outperformed the nanoporous MPL (20 mV reduction at 2.0 A/cm^2) due to increased pore access for oxygen and water.

The role of MPLs in improved catalyst utilization has also been investigated. As stated in **Section 4.1.2**, conductive catalyst supports are currently lacking. Thus, providing in-plane electrical conductivity between catalyst particles—and hence catalyst utilization—at low loadings will likely depend on the PTL surface. MPLs will likely be critical at low loadings, where poor in-plane electrical conductivity could result in fatal losses in catalyst utilization. Schuler et al.³⁹¹ constructed PTLs with MPLs of various pore sizes and studied their effect on catalyst utilization, albeit at higher loadings (2 mg cm^{-2}). The PTL structures are displayed in **Figure 17a**, and polarization curves in **Figure 17b**. The bulk PTL had a mean pore diameter of 27 mm (the single layer “SL” sample has no MPL), and MPL 1, 2, and 3 had mean pore diameters of $6, 11,$ and 13 mm , respectively. Overall, catalyst utilization for the MPL samples increased by a factor of $2.2\text{--}2.8$ compared to the SL sample, leading to $\sim 60\text{--}100 \text{ mV}$ reductions in overpotentials at 2 A/cm^2 . These voltage improvements were attributed to kinetic and mass transport improvements.

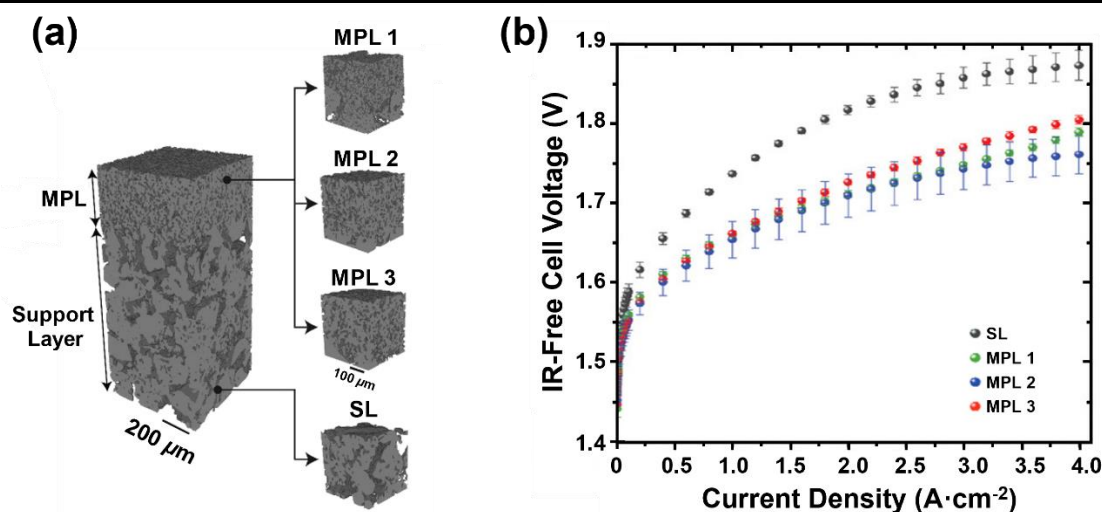


Figure 17. Effect of different MPL structures on performance. (a) The left figure portrays a generic PTL with MPL structure, while the right figures portray the MPL and single layer “SL” structures. MPL1, MPL 2, and MPL 3 differ in pore size and porosity. (b) Polarization curves for SL, MPL 1, MPL 2, and MPL 3 samples. Obtained using commercial Greenerity MEAs (Nafion N115 membrane) at 50°C and ambient pressure. Adapted with permission from ref.³⁹¹ Copyright 2019 Advanced Energy Materials.

4.4.7 Coatings

Coatings can help prevent surface oxidation and maintain CL-PTL conductivity. As such, coated PTLs have been shown to have increased performance and lower degradation rates than uncoated PTLs.^{341,392} Platinum is the most widely used coating material in commercial and academic research. Bernt et al.³⁹³ examined the impact of PTL coatings on cell performance and found that platinum coatings provide the necessary in-plane conductivity that low-loading MEAs severely lack. MPL usage at low loadings ($\sim 0.27 \text{ mg}_{\text{Ir}}/\text{cm}^2$) resulted in HFR reductions of over $15 \text{ m}\Omega \text{ cm}^2$ —resulting in $\sim 50 \text{ mV}$ improvement in cell voltage at 3 A/cm^2 . Rakousky et al.³⁹⁴ conducted a steady-state durability test for over 1000 hours and found that

a platinum-coated PTL had an 89% reduction in degradation rate ($108 \rightarrow 12$ $\mu\text{V}/\text{h}$) when compared to an uncoated PTL.

Coating degradation—caused predominately by metal dissolution and delamination—is heavily influenced by the application technique. Two widely used methods are electroplating and sputtering. Though electroplating is faster and simpler, it has uniformity and adhesion issues. In contrast, sputtering provides superior quality and precision (for thickness and material amount) but requires longer times and more costly equipment. As a result, coatings from sputtering exhibit prolonged durability even after prolonged usage. Regarding material selection, some studies suggest that iridium is a better coating than platinum because of improved stability and higher conductivity of its oxides.^{395,396} Moreover, since the iridium sits at the CL-PTL interface, it can offer OER assistance to the anode CL or even enable ionomer-free CLs. Thus, some argue that the benefits of iridium coatings outweigh the additional costs imparted to the stack. Alternative techniques for surface modification include using novel mixed metal oxide coatings³⁹⁷ and acid etching,³⁹⁸ both of which enhance corrosion resistance. Although these metal oxide coatings show promise, their long-term durability is yet to be shown. Acid etching results in titanium hydride subsurface layers on the PTL, which does not affect electrical conductivity, but enhances corrosion resistance by mitigating titanium oxide formation.³⁹⁹

4.5 Gas Diffusion Layer

The gas diffusion layer (GDL) is the cathode-side equivalent of a PTL and is responsible for thermal conduction, electrical conduction, and hydrogen removal. However, the cathode is a low-potential environment, and thus, corrosion is not a concern during operation. Due to similarities in property and function, PEMWE GDLs have generally been adopted from PEMFC GDLs with minimal modifications.^{400–403} Current industrial GDLs (such as those from AvCarb, Freudenberg, and Toray) are carbon-based and are usually made from felts, papers, or woven substrates. This combination of material composition and structure makes GDLs compressible, unlike PTLs. Thus far, few works exist that study the impact of the GDL on PEMWE performance, since unoptimized GDLs adapted from PEMFCs have proved more or less adequate. This ease of incorporation is due to low HER overpotentials (relative to OER) and high H_2 gas diffusion rates (relative to O_2 gas and water).

4.5.1 Compression

Since GDLs are compressible, they are responsible for stress absorption from membrane swelling and anode-related stress (since the PTL is incompressible). Compared to PEMFCs, the membranes used in PEMWEs are thicker, resulting in greater swelling and inducing additional stress on the GDL. Aside from stress absorption, GDL compression can also help prevent water flooding and promote BPP-GDL and GDL-CL electrical contact. Since hydrogen diffusivity is high, reductions in porosity due to compression should not significantly impact H_2 removal. However, GDLs can experience mechanical damage due to overcompression if the cell is not properly designed or gasketed.

On the other hand, GDL decompression can also be an issue, which can result if hydrogen at the cathode is being compressed. Such decompression can lead to increased contact resistances and, thus, greater ohmic overpotentials. Generally, the frame that hosts the anode and cathode components in PEMWEs is made to a specific thickness, but manufacturing complications sometimes prevent the components from taking up the desired thickness. To combat the possibility of decompression, a thick layer (up to several millimeters) of stiff carbon is added in the cell. For PEMFCs, optimal GDL compression sits at around 10-25%; this range provides effective transport and mechanical stability.⁴⁰³ However, optimal GDL compression for PEMWEs cannot be generalized as it will depend on the cell configuration (e.g., membrane thickness, flow field design, etc.).

4.5.2 Wettability

Since water diffuses through the membrane to the cathode via electroosmotic drag (**Figure 1**), a diluted hydrogen and water mixture ends up in the cathode outlet stream. Thus, hydrophobic GDLs are desirable as they mitigate water transport from the membrane into the outlet stream, resulting in pure hydrogen (also called “dry hydrogen”). Hydrophobicity can be attained using a hydrophobic base material or by incorporating a hydrophobic MPL into the GDL. PTFE is currently used as a hydrophobic additive, but it is not electrically conductive and thus contributes to higher GDL-BP interfacial contact resistances. Therefore, finding alternative materials to promote hydrophobicity is a research area.

4.5.3 Outlook

Although research is still needed for GDL optimization, existing designs derived from PEMFCs will be sufficient for GW-scale deployment. However, one note to consider is that PEMWE GDLs are usually carbonized at high temperatures in vacuum furnaces to eliminate impurities. As PEMWEs are more sensitive to cation contamination than PEMFCs. This additional step can increase manufacturing costs. However, this step may not be necessary as its impact on long-term performance has not yet been studied in the literature. Lastly, it should be considered that although GDL corrosion is typically not a concern, PEMWE shutdown events can lead to rapid carbon corrosion due to the presence of reverse currents. Thus, care should be taken to ensure that PEMWE durability is not affected by excessive corrosion.

4.6 Bipolar Plate

BPs are expensive due to material needs and the manufacturing process, contributing 15% to 50% of the total stack cost.^{404 405,406} BPs are responsible for mass transport (H_2O in; H_2 and O_2 out), electrical conduction, heat conduction, and mechanical support.^{404,407–409} Unlike PTLs, which are responsible for through-plane mass transport via diffusion, BPs contain flow fields responsible for in-plane mass transport via convection. As shown in **Figure 18a**, they are called “bipolar” because one side is in contact with the anode, and the other side is in contact with the cathode.⁴⁰⁷ For BPs to be effective, they must possess low contact resistance and efficiently transport water and gases. Furthermore, just like the PTL, BPs need to be made of a corrosive-resistant material that can withstand the acidic and oxidizing conditions of PEMWE cells.

The most common base materials for BPs are titanium and certain stainless steels (SS; e.g., SS316 and SS403), as shown in **Figure 18b i** and **ii**.^{404,407} Unlike for PEMFCs, graphite is only suitable for the cathode environment.⁴¹⁰ With the use of titanium or SS, coatings are needed to maintain low interfacial contact resistance with adjacent components. For SS specifically, the coating would also offer corrosion protection. Precious metals such as platinum, gold, and silver (**Figure 18b iii**) are typical coating materials, with platinum being the most common.^{405,407} To apply these coatings, electrical plating is conventionally used, but methods such as physical vapor deposition are in development.

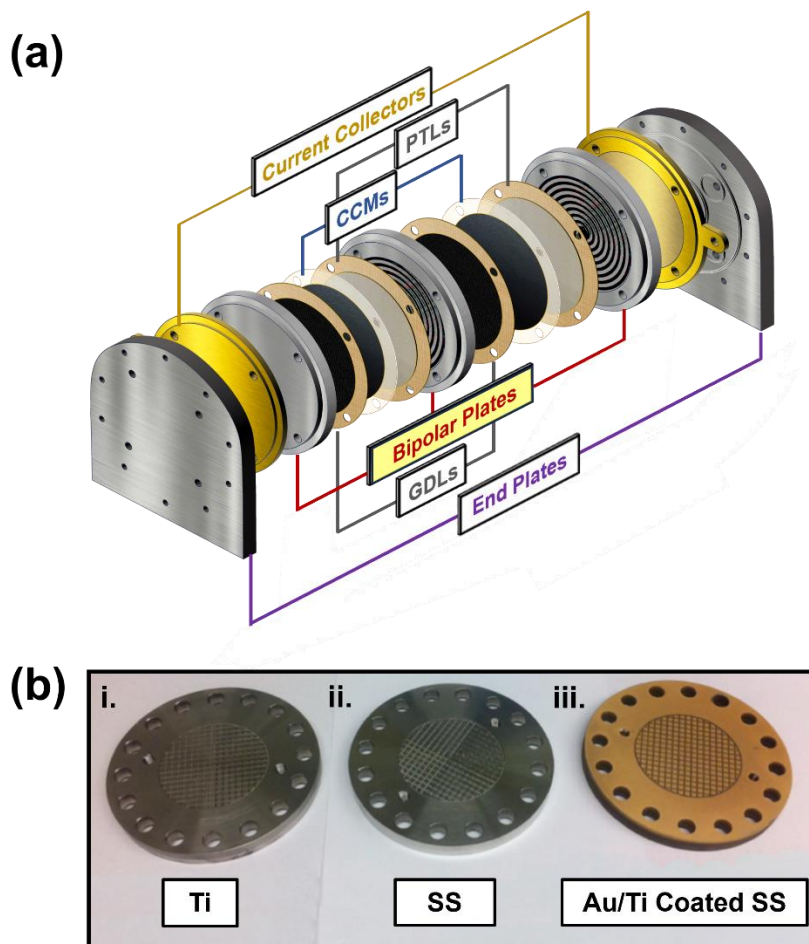


Figure 18. (a) General schematic of a PEMWE highlighting the placement of bipolar plates. (b) Examples of bipolar plates made of i) titanium, ii) stainless steel, and iii) stainless steel coated with a Au/Ti mix. Adapted with permission from ref.⁴¹¹ Copyright 2014 Journal of The Electrochemical Society.

Developments for BPs can leverage knowledge acquired from PEMFCs, but material limitations provide challenges.^{405,407} Fortunately, flow field designs and manufacturing techniques can more easily be adapted for PEMWE BPs. Overall, the primary limiting factor for scale-up is cost.

4.5.1 Flow Field

BP flow field design influences heat conduction and mass transport in PEMWEs. Since excess water flows throughout the cell for heat removal, flow fields must equally distribute this water to avoid hotspot formation. Additionally, the pressure drop across flow fields should be minimized to reduce pumping costs (which are associated with balance of stack).

Many methods can be employed to create a flow field, such as electroetching, stamping, electrical discharge machining, electrochemical machining, electroforming, and additive manufacturing.⁴¹² Three types of flow field structures exist: 1) Multi-layered, where layers of a metal mesh or screen are stacked on a flat base material (**Figure 19**). Pores of adjacent layers are offset to construct a 3-D hollow structure for transport. Pore sizes and the number of layers are critical design parameters. 2) Electroetched, where serpentine flow field channels are etched on a thick metal plate. This process simplifies the BP manufacturing process as only one component is needed, but suffers from high processing costs. 3) Stamped, where 3D flow structures are stamped onto a base. Stamping onto thin base materials (~0.5 mm)—especially with stainless steel—is common for PEMFC BPs.^{412–415} This stamping strategy has been shown to enhance mass transport.⁴¹⁶ Like the etched structure, assembly costs are reduced due to only needing one component. However, complications arise from difficulty controlling stress distortion.

Improper stress control during stamping could result in uneven surfaces and, ultimately, complications in stack sealing (especially under high pressures).

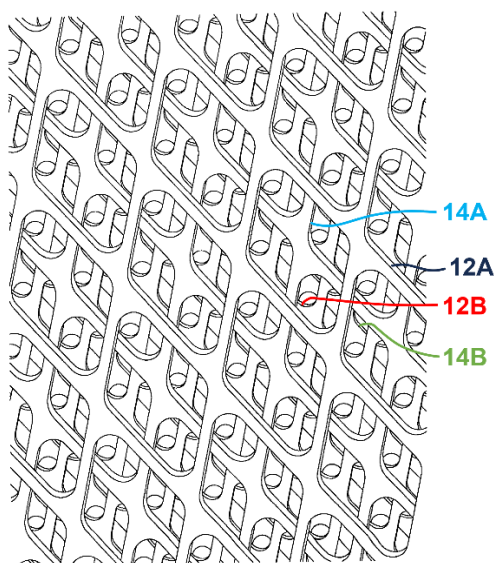


Figure 19. Example of a bipolar plate flow field design consisting of multi-layered metal screens. 12A, 12B, 14A, and 14B are separate layers with different diamond patterns used to form an optimized structure for water and gas flow. Adapted from ref.⁴¹⁷

Flow field performance is a strong candidate to be improved through modeling. Distribution profiles of oxygen, fluid velocity, pressure, temperature, and even current density can be constructed for various flow field types, as shown in **Figure 20**.^{418–421} These simulations require Multiphysics models that couples heat, mass, and charge transfer effects. Furthermore, two-phase flow effects through other cell components (like the PTL) must be included for comprehensiveness. Olesen et al.⁴²² modeled three circular interdigitated flow field variations. Reasonably, the regions with the highest temperature and oxygen content had the highest current densities. Interestingly, despite the similar overall performance, they found significant in-plane variation in temperature and current density between the three designs. Another study⁴²³ compared a novel interdigitated-jet hole flow field design with two common designs (standard interdigitated and single serpentine). They found the novel design to have more homogeneous water distribution and more effective gas removal, leading to more uniform current density and temperature profiles. Qing Wei et al.⁴²⁴ used a similar model to compare a novel mesh flow field with commonly used parallel flow fields. They found that the mesh structure enabled greater temperature and current density uniformity (**Figure 21**) as well as lower energy consumption.

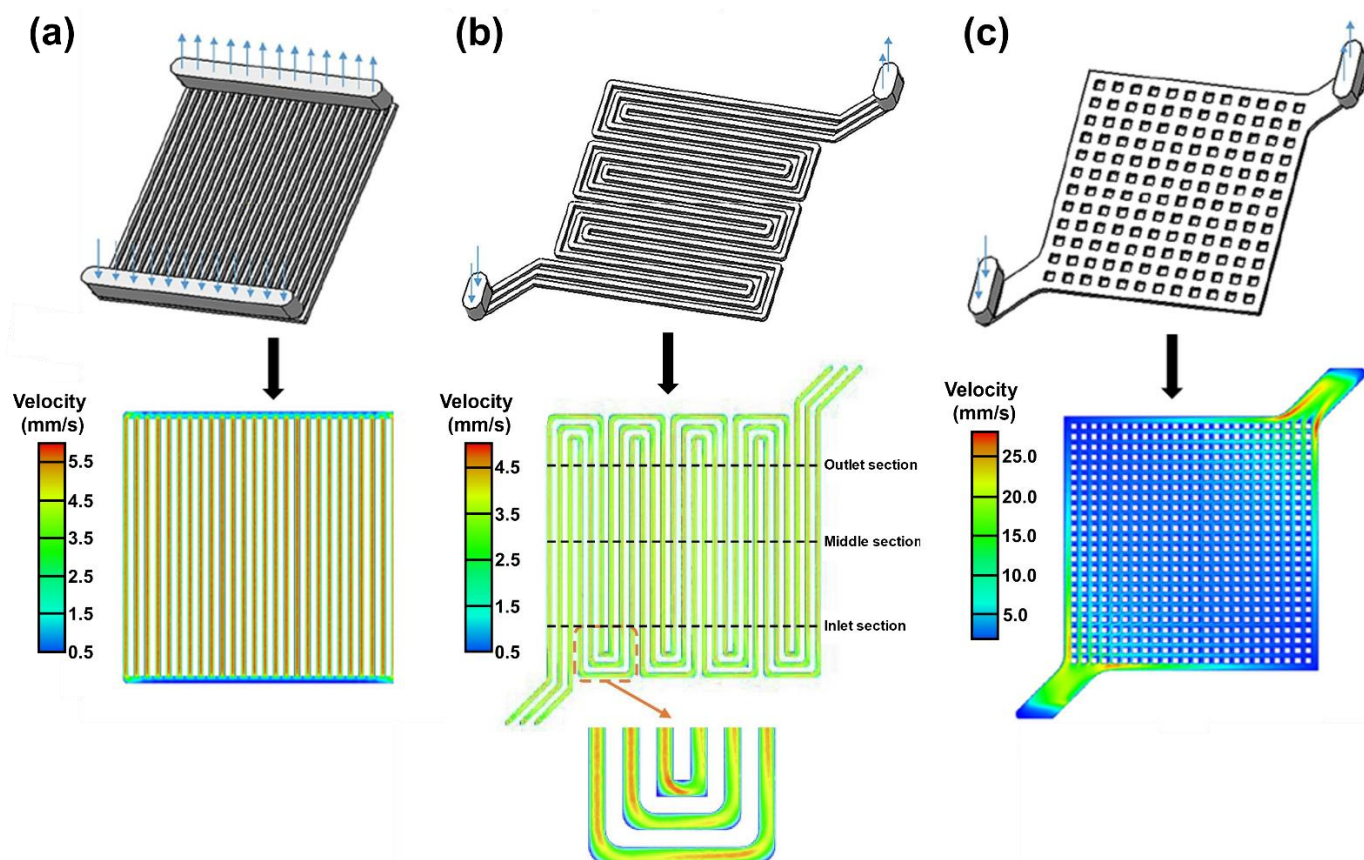


Figure 20. Examples of different flow field architectures and their resulting velocity profiles. (a) is a parallel flow field, (b) is a serpentine flow field, and (c) is a point flow field. Adapted with permission from ref.⁴²⁰ Copyright 2022 Applied Energy

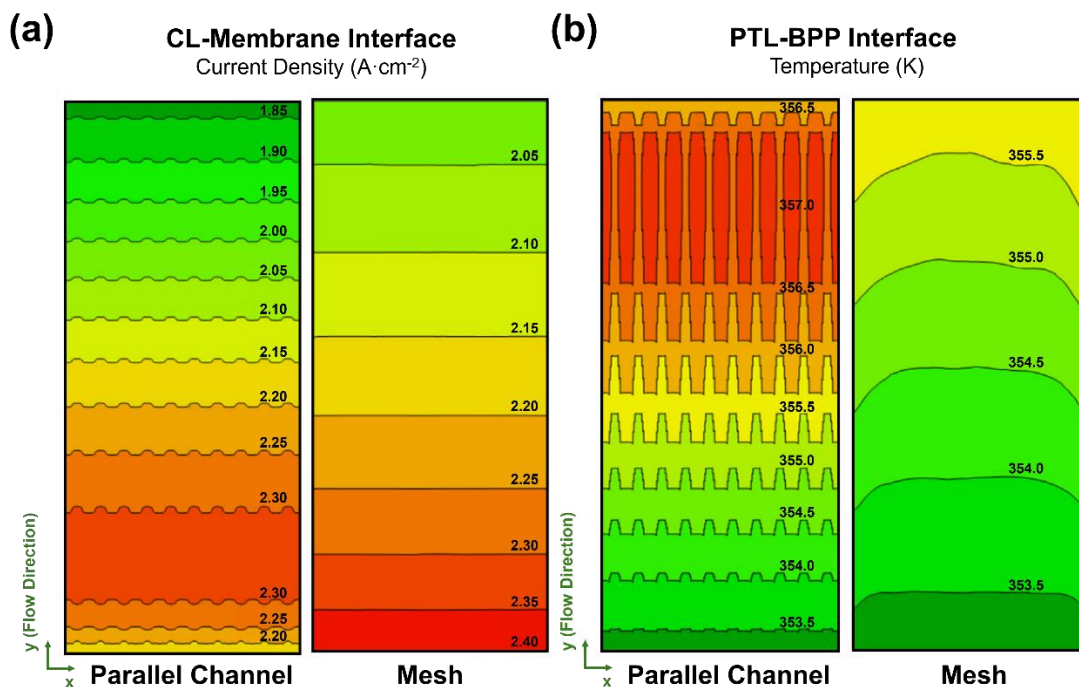


Figure 21. Performance comparison between a parallel channel flow field (Model A) and a mesh flow field (Model B). Contour plots showing (a) current density (A cm^{-2}) and (b) temperature (K) distributions. Adapted with permission from ref.⁴²⁴ Copyright 2023 International Journal of Hydrogen Energy.

4.5.2 Coatings

BP coatings must be stable in the highly corrosive environment of a PEMWE anode: oxygen-saturated, potentials $\sim 2\text{V}$, temperatures $\leq 80^\circ\text{C}$, and $\text{pH} \sim 0$ (**Figure 22**).^{410,425,426} Furthermore, they must also have low contact resistance, high through-plane electrical conductivity, good adherence to the BP, low cost, and scalable methods of deposition.^{425,426} Titanium BPs are usually coated with PGMs, as they meet the requirements above and can also prevent surface passivation.⁴¹⁰ If SS is used as the base material, the coating must also prevent ion leaching, which could lead to increased coating costs.^{425,426} However, SS is cheaper than titanium, and thus coated SS BPs could still be cheaper than coated titanium BPs.^{404,426} The method used to apply coatings could also influence costs; common methods include plasma, powder, and electrochemical deposition.⁴²⁷

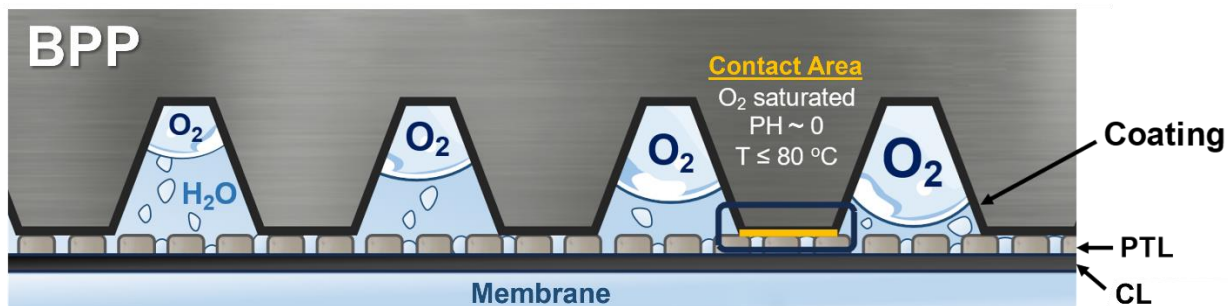


Figure 22. A schematic of the conditions relevant for BP design. Adapted with permission from ref. ⁴⁰⁶ Copyright 2016 Journal of Power Sources.

BP designs with novel coatings have also been demonstrated. Using a SS base, Gago et al.⁴²⁶ developed a two-layer coating consisting of a $60\ \mu\text{m}$ titanium layer and a $\sim 1\ \mu\text{m}$ platinum layer. The titanium provided corrosion resistance, while the platinum prevented the titanium from passivating. Future work dedicated to decreasing the titanium layer thickness can reduce coating costs.⁴²⁶ A different study by Rojas et al. ⁴²⁵ concluded that utilizing a SS BP with a multi-layer Ti/TiN coating can reduce manufacturing costs by 90% compared to titanium BPs. However, further work must be completed to make this technique more scalable.

4.5.3 Hydrogen Embrittlement

Hydrogen absorption can occur on the cathode side of BPs due to high hydrogen pressures and radical generation from the HER, weakening the mechanical integrity of the BP and leading to a phenomenon known as “hydrogen embrittlement.”⁴²⁸ Titanium BPs can suffer mechanical failure with hydrogen concentrations over 8,000 ppm,⁴²⁹ and it was reported that such plates could absorb over 1000 ppm in 500 hours of operation (meaning that stack failure could occur in just 4,000 hours).⁴³⁰ Even using platinum coatings, hydrogen embrittlement can still occur since hydrogen can absorb in the platinum and form Pt-H bonds.⁴³¹

Two approaches to prevent hydrogen embrittlement are applying a gold coating or a pinhole-free carbon coating to prevent direct contact with hydrogen. Gold can help prevent hydrogen absorption, but due to its high diffusion coefficient, a thick coating ($>200\ \text{nm}$) is required to meet lifetime requirements. Such high precious metal usage presents cost issues. Another approach is to have a pinhole-free carbon coating, which has been shown to reduce hydrogen absorption.⁴³⁰ However, challenges lie in costs and processing difficulties associated with pinhole-free manufacturing. Lastly, a more roundabout strategy would be to allow the titanium surface to form surface oxides, which would prevent hydrogen absorption, and then partially cover the surface with gold dots to enable adequate electrical conductivity (**Figure 23**).⁴³² It was shown that surface coverage of less than 10% with gold dots would meet contact resistance requirements. Overall, hydrogen embrittlement is still a significant issue and will require developing effective coating materials and scalable coating methods.⁴³³

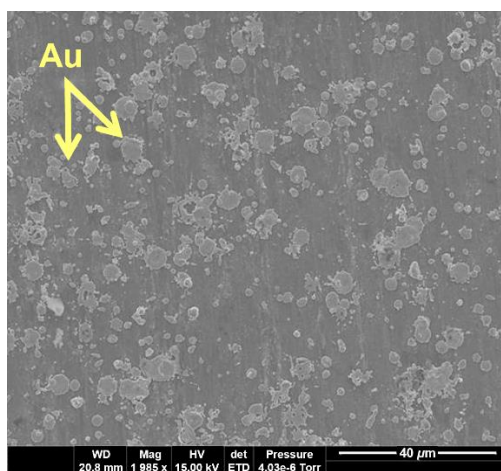


Figure 23. SEM photo of a titanium BP covered with gold dots to improve interfacial electrical contact. The uncovered surface is composed of a thick titanium oxide layer to prevent hydrogen absorption.

4.5.4 Outlook

Current BPs are made of titanium coated with platinum, constituting up to 50% of stack costs. For PEMWE scale-up, the costs of base and coating materials must be reduced. For the base, stainless steel is an attractive option; however, a coating that is inexpensive and durable (towards corrosion and ion leaching) is required. Platinum can still be used as a coating, but loadings—which currently sit at $\sim 1\text{--}5\text{ mg/cm}^2$ —should be reduced to decrease costs.⁴³⁴ Of course, platinum-free coatings are ideal for cost reduction, but further work into effectiveness and scalability is required. Lastly, hydrogen embrittlement presents a major durability issue and needs to be addressed with novel coating strategies.

5 AST Proposals

ASTs are durability testing protocols that enhance the degradation of components under industrially relevant conditions. These tests are needed to determine system lifetimes without numerous years of testing. Furthermore, ASTs can help us understand degradation under different operating regimes, which may be useful in future scenarios. For example, although current PEMWEs operate at constant loads to minimize degradation, techno-economic analysis indicates that dynamic operation following cheap renewable electricity (i.e., “load following”) will be critical to producing low-cost hydrogen.⁴³⁵ While numerous papers have examined PEMWE durability,^{436–439} no standardized AST protocols exist.

The U.S. DoE has published various standardized ASTs for PEMFCs, including widely used component-level tests, as well as rarely used MEA-level tests for overall system lifetime predictions.⁴⁴⁰ Their “Research, Development, and Demonstration Plan”⁴⁴⁰ lists ASTs for electrocatalysts, catalyst supports, membrane chemical stability, membrane mechanical stability, membrane chemical/mechanical stability, and drive cycle durability. While the first five tests target specific components and have been widely used in the literature, the last AST (“drive cycle durability protocol”) targets overall system durability under light-duty operating conditions and has rarely been reported.^{441,442} Since lifetime predictions are highly dependent on the exact duty cycle used in any particular application, it is more common for this type of AST to be specifically developed by original equipment manufacturers (OEMs).

Partnerships between OEMs and researchers can facilitate durability improvements. OEMs can provide researchers with end-of-life materials and data pointing to specific failure modes, so that efforts are not misappropriated to irrelevant durability concerns. At the same time, researchers can use their resources to understand the fundamental issues governing degradation. Given the criticality of meeting lifetime targets, researchers must prioritize the expansion of testing capacities (both ex-situ and operando) to probe durability topics and perform ASTs.

Although less standardized, PEMWE ASTs have been proposed for the anode, cathode, and overall system. The EU published a harmonized report on electrolyzer testing and found the “key stressors” for PEMWE durability to be temperature, pressure, water flow rate, and water quality.⁴⁴³ Using these four stressors, eight ASTs were developed for anode and cathode degradation. Two overall system ASTs were also proposed: a “reactive” test with 1 min step durations and a “flexible” test with 15 min step durations (each step was a current hold, ranging from 25-200% of nominal current density). Moreover, Aßmann et al.⁴⁴⁴ proposed overall system ASTs based on different operating modes: low load, high load, dynamic load, and startup/shutdown. While a handful of overall system ASTs have been proposed, component-specific tests are lacking. Here, we propose ASTs for each cell component, summarized in **Table 3**.

Lastly, it should be noted that component-specific ASTs could produce unwanted stress on other components, leading to seemingly contradictory results. For example, a study by Alia et al.¹³⁷ found that dynamic operation causes more degradation than constant operation, while another study by Frensch et al.²⁹⁴ reported the opposite. However, performance losses were attributed to kinetic overpotentials in the first study and ohmic overpotentials in the second. Thus, care should be taken to meticulously probe the cause of electrochemical performance loss in an AST (e.g., catalyst dissolution, membrane thinning, PTL/BP passivation, etc.), regardless of which component is targeted.

Table 3. Proposed component-specific accelerated stress tests (ASTs) for PEMWEs

Component	Proposed AST	Specifier
Anode catalyst	1.45 V – 2.0 V cycling	Standard Operation
	0.5 V – 2.0 V cycling	Start/Stop Operation
Cathode catalyst	0.5 V – 2.0 V cycling	/
Membrane	OCV in H ₂ /air	Chemical Stability
	1-200 bar cycling in air/air	Mechanical Stability
	1-200 bar cycling in H ₂ /air	Overall Stability
PTL	2.0V hold	/
GDL	Air/H ₂ cathode cycling	/
Bipolar Plate	Ex-situ 2.0 V hold	Ex-situ
	90°C operation	In-situ

5.1 Anode Catalyst Layer

PEMFC catalyst ASTs involve cycling between two potentials where platinum is in different oxidation states (metallic platinum and platinum oxide/hydroxide) and thus exhibits vastly different dissolution rates.⁴⁴⁵ This cycling results in catalyst agglomeration and loss of electrochemically active surface area.⁴⁴¹ In addition to potential cycling, temperature and relative humidity (RH) also accelerate degradation.⁴⁴⁶ Thus, the AST (for the cathode catalyst) is 0.6V-0.95V cycling (2.5s holds) at 80°C and 100% RH. Furthermore, the test is performed in an H₂/N₂ environment to prevent ionomer degradation.

PEMWE anode catalyst ASTs can be designed with a similar potential cycling strategy, as transitions to lower voltages and OCV conditions were shown to be highly degrading (**Section 4.1.3**). To assess durability representative of standard operation, 1.45V-2.0V cycling can be used; for durability representative of shutdown/startup operation, 0.5V-2.0V cycling can be used. For both tests, an elevated temperature of 80-90°C can be used, and the potential hold time can be adjusted between 10-30s. This time window allows for rapid cycling but also provides adequate time for iridium to undergo oxidation state changes. Electrochemical performance at kinetically-dominated regions (i.e., at low current densities) can be used to track catalyst degradation.

5.2 Cathode Catalyst Layer

The durability of the cathode CL has drawn some attention regarding start/stop operation. Grigoriev et al.⁴⁴⁷ reported significant platinum dissolution attributed to the 600+ shutdown events in their 4000h test. During shutdown, reverse currents can occur (i.e., the system works as a fuel cell) and trigger mechanisms of platinum dissolution until all electrolysis gases are exhausted (the authors suggest purging the anode and cathode gases as mitigation strategies).⁴⁴⁷ Voronova et al.⁴⁴⁸ also observed this degradation mechanism, as they found performance losses when cycling between 1.4-2.2V, but not when cycling between 1.5-2.2V.

Since shutdown conditions impart the greatest stress on the cathode catalyst, we propose the same test as the startup/shutdown AST for the anode catalyst: 0.5-2.0 V cycling. While OCV-load cycling has been seen in the literature for cathode degradation,⁴⁴⁹ we do not recommend incorporating OCV conditions in standardized protocols as reproducibility will likely be unsatisfactory. The nature of OCV conditions (i.e., how quickly the potential decays, what value the potential will decay to, etc.) will depend on the rate of hydrogen crossover, which can vary significantly with membrane thickness and chemistry.

5.3 Membrane

Membrane degradation has been identified as one of the main failure mechanisms for PEMWEs^{450–452} and was discussed extensively in **Section 4.3.4**. A summary of the two degradation modes—chemical and mechanical—will be presented here. Chemical degradation can lead to membrane thinning²¹³ and occurs when radical species attack the PFSA ionomer (**Figure 11**). The rate of degradation can be quantified by measuring FRRs. A model by Chandesris et al.⁴⁵³ found that temperature accelerates FRRs and that higher rates are seen at the cathode. Moreover, current cycling was reported to cause increased FRRs compared to steady-state operation.⁴⁵⁴ Mechanical degradation as a failure mode has been demonstrated with thin N211 membranes (~25 μm).⁴⁵⁵ Membranes can experience mechanical stress through delamination or the formation of hotspots. It has been reported that thicker membranes penetrate 19 to 45 μm into the PTL, with factors such as membrane thickness and PTL porosity influencing the extent of penetration.⁴⁵⁶ Furthermore, flow field designs that lead to non-uniform $\text{H}_2\text{O}/\text{O}_2$ distributions across the cell can result in membrane hotspots that accelerate degradation.⁴⁵⁷

Three membrane ASTs for PEMFCs currently exist: OCV hold in H_2/air (for chemical degradation), RH cycling (for mechanical degradation), and RH cycling in H_2/air (for combined chemical/mechanical degradation).^{442,458} Since mechanisms of membrane degradation are similar between PEMFCs and PEMWEs, these ASTs can be adapted for PEMWEs. RH cycling can be replaced by pressure cycling, which is more relevant to electrolyzer operation. For chemical degradation, an OCV hold in H_2/air at 90°C and 30% RH can be performed. For mechanical degradation, pressure cycling between 1-200 bar (which covers relevant pressure ranges) in air/air at 90°C can be done. For a combined chemical and mechanical test, the pressure cycling can be performed in H_2/air . Lastly, a dual membrane and anode catalyst AST can be created by combining pressure cycling with the startup/shutdown anode catalyst AST, resulting in cycling between 1 bar/0.5 V and 200 bar/2.0 V. Catalyst degradation can be tracked via polarization curves, while membrane degradation can be tracked via HFR changes or H_2 crossover rates.

5.4 Porous Transport Layer

As catalyst utilization/degradation heavily depends on the CL-PTL interface, developing durable PTLs will be critical. Surface passivation is a primary PTL degradation mechanism, leading to increased kinetic and ohmic overpotentials. Coatings can help mitigate passivation, but the coating itself is subject to degradation in the form of dissolution and delamination. Thus, since PTL durability is tied to surface passivation, a constant high voltage load of 2.0V would be an appropriate AST. It should be noted that PTL degradation is intimately tied with catalyst degradation, so both should be evaluated in conjunction.

5.5 Gas Diffusion Layer

GDLs have been shown to corrode under the oxidizing conditions of PEMFC anodes,^{459,460} but their corrosion under the reducing conditions of PEMWE cathodes is typically not a concern. However, PEMWE shutdown events can lead to reverse currents and trigger carbon oxidation. Thus, if frequent shutdown events are implemented in an operating strategy, care should be taken to ensure that GDL corrosion does not significantly affect system durability. For an AST, since the GDL environment does not experience oxidizing conditions during regular operation, cathode cycling between oxidizing (air) and reducing (H₂) environments could be used—as proposed by Perry et al.⁴⁶⁰

5.6 Bipolar Plates

Despite the BPP's importance in durability and cost considerations, BPP ASTs have not been proposed for PEMFCs or PEMWEs. However, the U.S. DoE has published targets for PEMFC BPPs, including H₂ permeation and corrosion resistance.⁴⁶¹ To quantify corrosion resistance, ex-situ tests are performed where corrosion currents are measured via chronoamperometry at relevant potentials in a simulated PEMFC environment—pH 3 with 0.1 ppm of fluoride anions (to represent membrane degradation products). It may seem logical to perform similar measurements for PEMWE BPs, but currents are dominated by H₂ and O₂ evolution at relevant potentials (i.e., above 1.5V or below 0V). Nevertheless, an ex-situ test can still be performed to examine ion leaching, H₂ permeation, and contact resistance. The test can be performed at 90°C, pH 3, 0.1 ppm HF, and +2.0V vs. SHE (for anode conditions) or -0.2V vs. SHE (for cathode conditions).

However, it should be stated that stack OEMs prefer to see in-situ durability data for BPPs, which can be highly time-consuming to obtain. OEMs typically use their own protocols based on targeted operating conditions, but for a general in-situ AST, we propose an elevated operating temperature of 90°C to accelerate corrosion and hydrogen embrittlement.

6 Industrial Perspectives

6.1 Nel Hydrogen

Nel Hydrogen is a global leader in deployed water electrolyzer systems, commercially selling liquid alkaline and proton exchange membrane products worldwide. While anticipating the hydrogen economy to take shape, Nel Hydrogen has been fortunate to sell electrolyzers for industrial applications, such as generator turbines (cooling gas), weather balloons (filling), heat-treatment furnaces (atmosphere control), and other applications requiring a reducing environment. Along with a focus on continuous durability and electrical efficiency improvements, Nel has aimed to scale up devices to meet emerging industrial and energy storage markets for green hydrogen. As shown in **Figure 24**, significant opportunities exist for green hydrogen to decarbonize existing industrial processes.

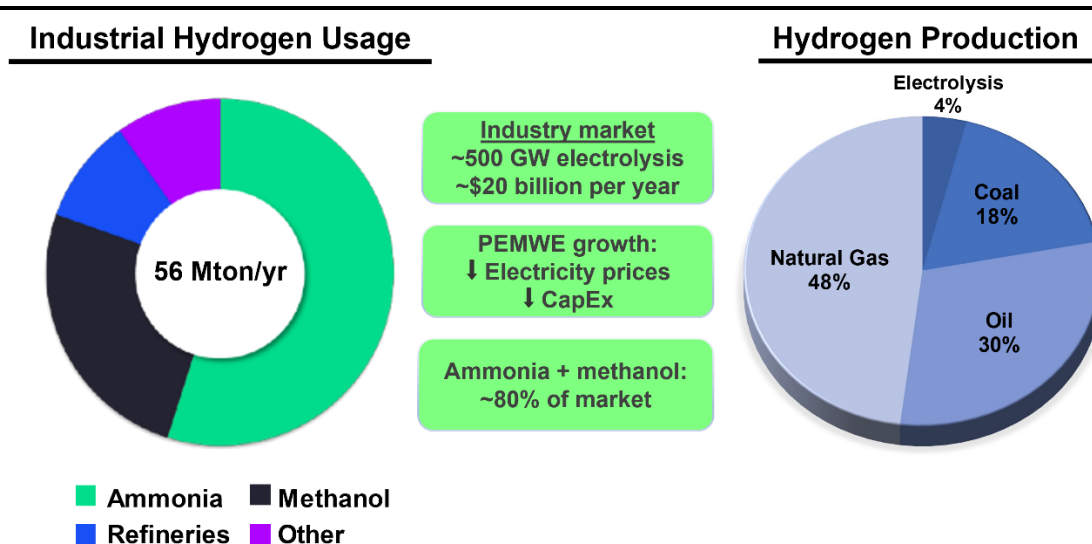


Figure 24. Overview of the current industrial H₂ market and methods for H₂ production.

Nel has supported the hydrogen economy by developing a broad portfolio of systems, spanning kW to hundreds of MWs. The scale-up of equipment and devices not only enables market penetration into areas requiring tons of H₂ generation per day, but also serves to reduce CapEx. Such CapEx reductions are critical to meet green H₂ cost targets—such as \$1/kg by 2031 (U.S. DOE)—and compete with H₂ produced through SMR. **Figure 25a** depicts the two main pathways for cost reduction: 1) technological development, which includes factors such as lowering catalyst loadings and using thinner membranes, and 2) scale-up for large-scale production. Technological development offers significant opportunities for cost reductions, but economic returns gradually flatten as advancements are implemented. Fortunately, scale-up can provide further cost reductions, approximated by the rule of “six-tenths” (**Figure 25b**). According to this rule, for a given cost of a PEMWE system (C_A), if manufacturing capacities increase by a factor of X , the final cost (C_B) decreases by a factor of $X^{0.6}$.

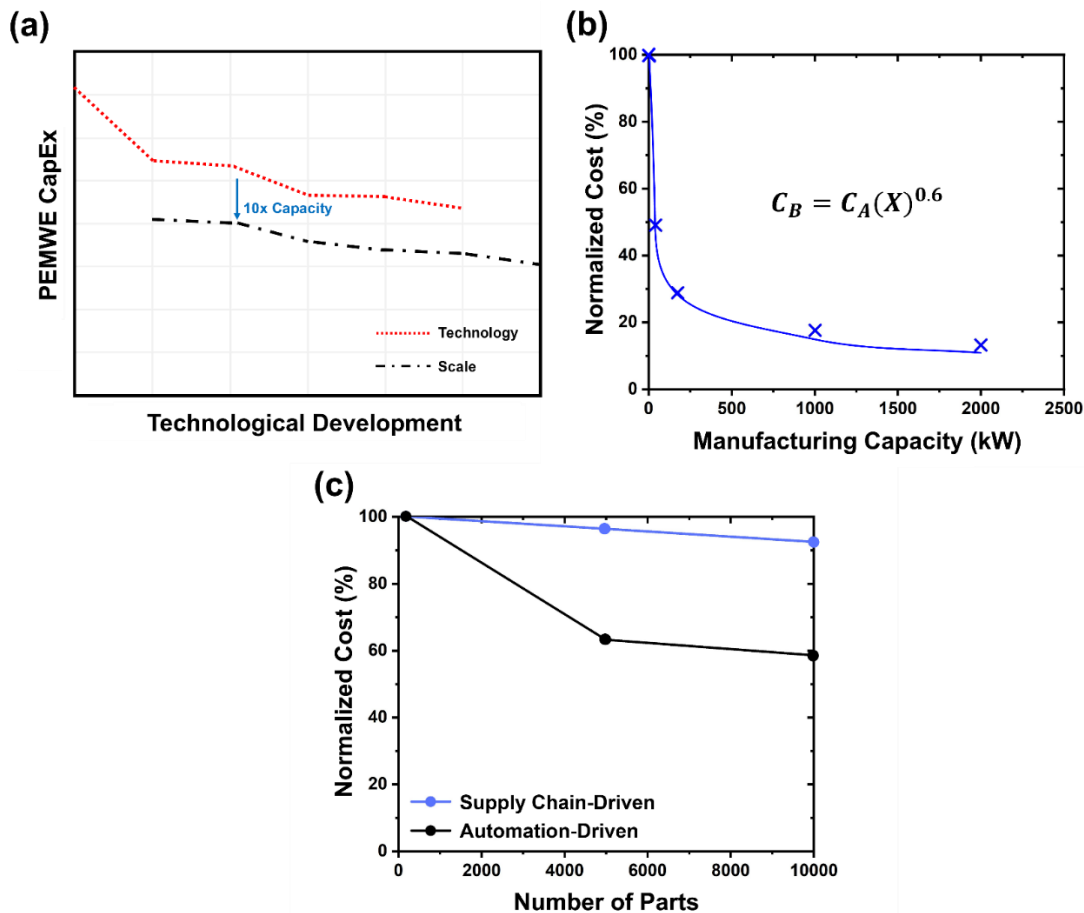


Figure 25. Graphically portrayals of cost reduction concepts. (a) Cost reductions through technology improvement and scale-up. (b) Rule of six tenths as applied to electrolyzer systems. C is Normalized Cost (%) while S is System Output (kW). (c) Cost reductions via volume discounts and automation.

To enable scale-up, Nel has continuously worked to increase stack size in active area and cell count. Such increases have not always been an easy task due to limitations in the existing supply chain. The available form factor of existing components has often been a constraint dictating how large these systems can go, with stack configurations frequently forced back into design space. However, with the scale-up of green hydrogen, this limitation is beginning to diminish. Thus, suppliers have recognized this time as an opportunity to expand their businesses and are open to developing their own manufacturing methods to address larger scales. This increase in supplier engagement has additional benefits related to cost: 1) the formation of supply chain agreements, which contribute to large orders and volume discounts, and 2)

direct engagement in the design phase from suppliers, which leads to more conscious stack design tailored for automation. These concepts are illustrated in **Figure 25c**, with the blue plot showing cost reductions via supply chain agreements and the black plot showing cost reductions via automation, which includes the most rudimentary steps (such as gasket cutting) all the way to stack assembly.

As renewable electricity costs continue to decline, the acceptance of PEMWE systems is becoming more widespread and will continue to build momentum. The challenge is for us to utilize technological advancements, increase production scales, obtain supplier engagement, and implement high-volume manufacturing processes to position electrolysis as a cost-effective solution to help global decarbonization.

6.2 Undisclosed Company

Understanding the broader context of a hydrogen economy is essential when approaching PEMWE development. This includes aspects such as large-scale H_2 production, storage, and distribution. A comprehensive pioneering initiative should involve green H_2 production in an interconnected H_2 -fueled ecosystem; a model of this concept is illustrated in **Figure 26**. In such a system, green H_2 generated by PEMWEs is distributed across a connected infrastructure to power buildings, fuel both light and heavy-duty fuel cell vehicles (at H_2 refueling stations), and decarbonize industrial plants. Currently, since there is no significant market for green H_2 , the demonstration and deployment of PEMWEs in an ecosystem would help build supply and demand. These demonstrations—such as the one in Fukushima⁴⁶²—can provide valuable information for understanding the technical feasibility and economic viability of green H_2 . Additionally, such a demonstration serves as an invaluable testbed for assessing performance limitations and degradation over time, providing critical real-world data to refine PEMWE technology.

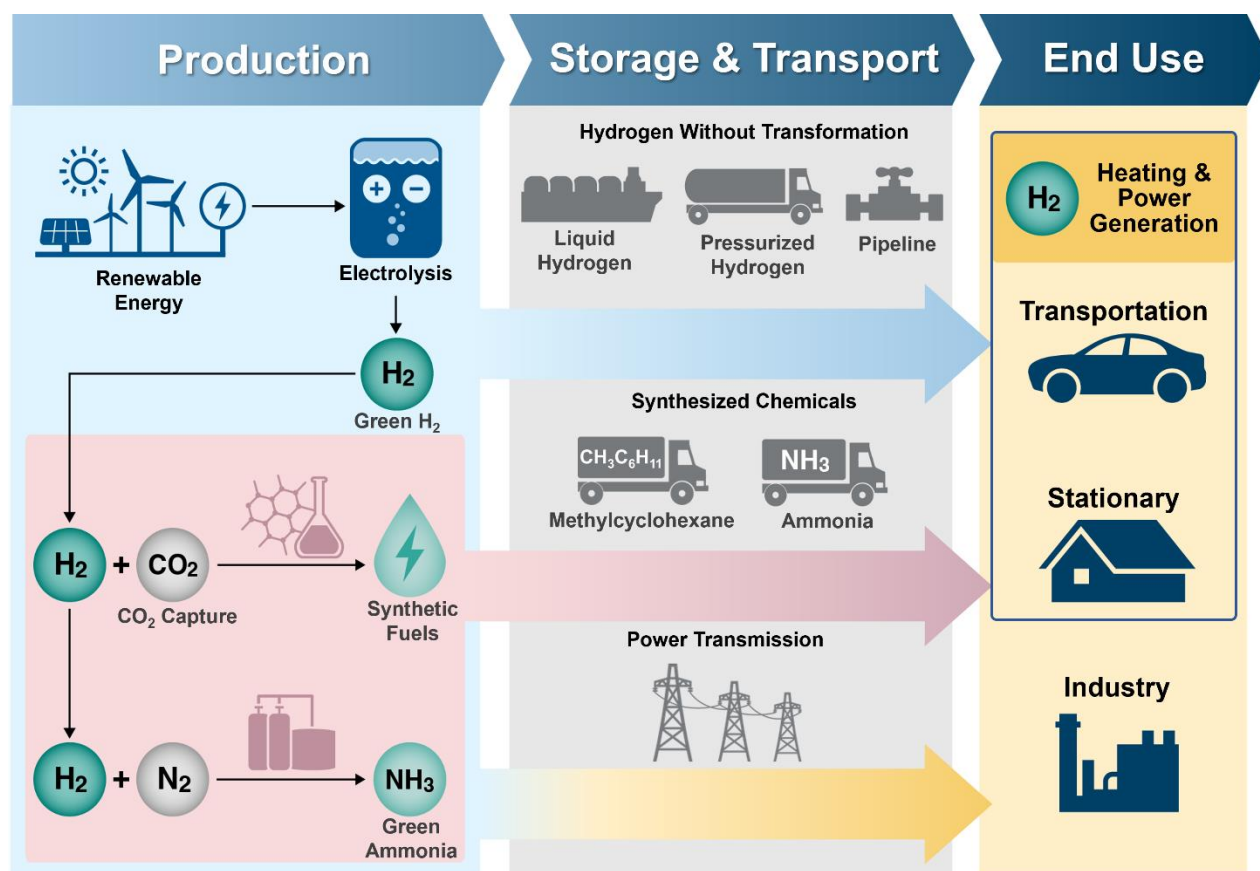


Figure 26. Schematic of a green hydrogen ecosystem. Green hydrogen production, storage, transportation, and end use depicted.

The primary challenge in green H₂ is reducing production costs.^{463,464} In addition to achieving economies of scale, there are multiple pathways to reduce costs: low-cost electricity coupling, material innovation, and PEMFC synergy. Firstly, electricity prices are the single largest driver for overall production costs. Therefore, it is imperative to couple PEMWEs with low-cost electricity from renewable energy sources like wind and solar. Thus, dynamic operation of PEMWE systems needs to be optimized for minimal performance degradation.

Secondly, material innovation plays a pivotal role in cost reduction. The reliance on iridium, one of Earth's rarest elements, presents a significant challenge. Developing catalyst materials with increased activity and stability is essential to meet efficiency, lifetime, and catalyst loading targets. Current commercial PEMWEs use a loading of $\sim 3 \text{ mg}_{\text{Ir}} \text{ cm}^{-2}$; reducing this to $0.1 \text{ mg}_{\text{Ir}} \text{ cm}^{-2}$ or less is critical to make large-scale deployment economically feasible.⁴⁶⁵ Furthermore, manufacturing MEAs with such low loadings presents significant challenges with existing coating methods, such as doctor blade coating or slot die coating. Thus, innovations in catalyst design and large-scale CL fabrication techniques are imperative.

Thirdly, synergy with already established fuel cell technologies could also reduce costs. Many components in PEMWEs—including the cathode catalyst, membrane, and BPP—are common with those used in PEMFCs. A strategic synergy between these two technologies can accelerate the initial development phase and capitalize on economies of scale, leading to significant reductions in investment costs for PEMWEs. Moreover, the shared components can enable greater cost efficiencies—particularly in mass production—due to common supply chains and manufacturing processes.⁴⁶⁶

With combined efforts in material innovation, cost reduction, and real-world demonstrations, GW-scale PEMWE deployment can be achieved. Ultimately, demonstrating the technical feasibility and economic viability of a green H₂ ecosystem is critical for PEMWE manufacturers to establish a sustainable business.

6.3 Plug Power

PEMWE devices are rapidly moving from niche applications to projects greater than 100 MW. The U.S. has pledged \$9.5 billion to hydrogen production hubs and electrolyzer research, while Europe is looking to add 40 GW of green hydrogen by 2030. This scale-up brings with it several challenges. First, the supply chain is not currently set up for these developments, and rapid production scaling could put pressure on precious PGM materials used as catalysts. The maturity of regulations, site permitting, and codes vary wildly by location, leading to difficulty in design standardization for global deployment. An influx of interest and money has led to thousands of new entrants, few with the experience and history necessary for large-scale projects. Lastly, with all this considered, safety must be at the forefront of system deployments.

Scaling up PEMWE systems requires a consistent supply of critical components, such as catalysts, membrane materials, and PTLs. Limited production capacities of these components can lead to bottlenecks, delays, and increased costs. Additionally, the demand for rare catalyst materials, especially iridium, can result in supply chain vulnerabilities. Fortunately, multiple studies have shown that iridium usage is low-risk, and PEMWEs can reach the terawatt level with decreased loadings, increased current densities, and iridium recycling.^{467,468} Another concern is the PFSA materials necessary for the membrane, binders, and gasketing. Bans on such materials are under consideration in both the E.U. and the U.S. Though there is active research in non-PFSA materials, nothing to date has shown adequate durability under realistic conditions ($>50,000 \text{ h}$ at $>60^\circ\text{C}$).

Large-scale PEMWE projects must adhere to various codes and standards to ensure operational reliability and safety. These codes cover equipment design, installation, operation, maintenance, and emergency response. Such regulations are most mature in Europe, where stacks usually need to be ATEX certified (*Atmosphère Explosible*; “Explosive Atmospheres”; ensures safe use in explosive environments) and must obtain a CE mark (*conformité européenne*; “European Conformity”; affirms conformity with health, safety, and environmental standards). The interior of a stack is generally, and rightfully, considered an explosive environment, requiring ATEX certification. Afterward, to obtain a CE mark, the stack must meet the “Pressure Equipment Directive,” demonstrating that it can withstand a multiple of the maximum allowed working pressure. This process requires additional design, testing, and manufacturing

certifications from notified bodies. In the U.S., it is generally left to local regulatory bodies who often rely on ASME (American Society of Mechanical Engineers) standards. Predictably, this flexibility leads to wildly varying permitting needs and timeframes. Only recently was a unified path being created for an electrolyzer certification scheme—called “U stamp”—for stack quality and engineering requirements, developed through an ASME case code.

Scaling up PEMWE systems involves compliance with local, regional, and national regulations. Obtaining hydrogen production, storage, and transportation permits can be complex and time-consuming. A good starting point for understanding the hundreds of codes and requirements from the stack to the system level is reviewing the guidelines for the U.K. and the U.S. state of California.^{469,470} Zoning restrictions, environmental impact assessments, and public safety must be considered. Collaborative efforts between industry stakeholders and regulatory bodies are necessary to streamline permitting procedures and ensure efficient project execution. Commonality is critical to bringing hydrogen generation to multiple locations, as regulatory uncertainty can defeat promising projects before they begin. R&D is also vital for scale-up: innovations in catalyst loading reduction, non-PFSA ionomers, energy efficiency, system design, safety measures, and supply chain optimization can collectively alleviate the difficulties associated with large-scale implementation. Public-private partnerships, academic collaborations, and government support are actively being pursued in these areas, with targets established for cost and performance to bridge efforts together.²⁴²

As the scale of PEMWE systems increases, so do safety concerns. Hydrogen, though a clean fuel, is highly flammable and poses explosion risks. Inadequate handling and storage of hydrogen can result in catastrophic accidents. Proper personnel training, stringent safety protocols, leak detection systems, and robust ventilation infrastructure are imperative, especially in large-scale installations. Although codes and standards exist for stacks and systems, they are often overlooked due to their immaturity or inconsistency between locations. Safe stack operation requires constant monitoring of voltage, water flow, temperature, pressure, and—most importantly— H_2 content (on the anode side). However, even with constant system monitoring, stack designers must plan for worst-case scenarios. As an example, Plug Power tests all system components for catastrophic failure; **Figure 27** shows the time-evolved progression of one of these tests, where a 1 MW stack and its anode outlet are subjected to worse-case H_2/O_2 stoichiometries that could result during a complete failure (i.e., membrane rupture). The H_2/O_2 gas mixture is then detonated, and the system is observed to ensure that physical explosion risks are contained and that the stack maintains its physical integrity. To avoid premature ignition, one must remove the catalysts in the stack before testing. Similar tests are done on the H_2O/O_2 separator, which is generally the system component with the most significant potential for explosive damage.

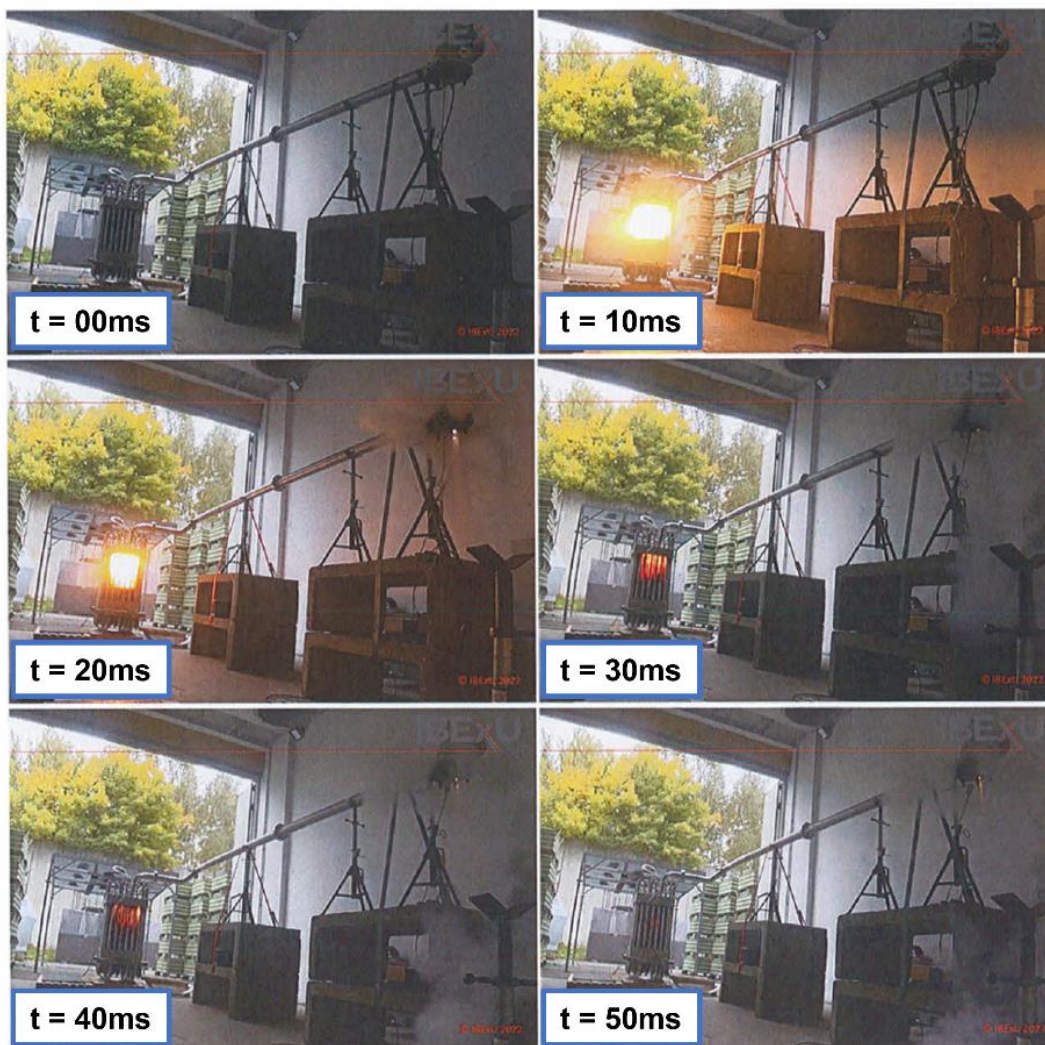


Figure 27. Testing of stack detonation under worst-case H_2/O_2 stoichiometric conditions. Test ensures that the stack contains the explosion and that no shrapnel is generated upon failure.

Scaling up PEMWEs holds the key to unlocking hydrogen's potential as a clean energy carrier. However, challenges related to supply chain management, safety considerations, permitting procedures, and adherence to codes and standards must be effectively addressed. Collaboration among stakeholders, regulatory bodies, researchers, and industrial companies is essential to overcome these challenges and pave the way for a sustainable and efficient GW-scale PEMWE industry.

7 Outlook

In the face of the dire consequences of climate change, much optimism exists for PEMWEs. To reach net zero emissions, deployment must scale up tremendously from 0.3 GW to 220 GW by 2030. This magnitude of growth will require concerted global efforts in every facet of PEMWE deployment—from fundamental research and component improvements to business strategies and federal policies. This section will provide a roadmap for the path forward.

7.1 Technical Targets

Overall PEMWE technical targets are proposed in

Table 4. These values are based on a compilation of reports from numerous agencies and academic papers from across the globe, with representation from North America (U.S. DoE),²⁴² South America,⁴⁷¹ Africa,⁴⁷² Asia,^{473–475} Europe (E.U. Clean Hydrogen Partnership, FCHJU),^{476–480} and international agencies (IRENA).¹⁴¹ Once targets were proposed, they were corroborated by leading representatives in industry.

Table 4. Technical targets for PEM Water Electrolysis

Parameter	Units	2024 Status	2030 Target	2050 Target	R&D Focus	Importance
System CapEx	\$/kW	1000	700	200	BoP	Med
Stack CapEx	\$/kW	400	150	50	MEA, PTL, BPP	High
H ₂ Production Cost	\$/kg	5	1.5	0.5	MEA	High
Lifetime	1000 h	60	80	100	MEA, PTL	Med
Degradation	%/1000 h	25	15	10	MEA, PTL	Med
System Efficiency	% LHV	65	70	75	BoP	Med
Stack Efficiency	% LHV	70	75	80	MEA	Med
Current Density (Nominal)	A cm ⁻²	2	3	5	MEA	High
Voltage (Nominal)	V	2	1.8	1.7	PEM	Med
Anode Loading	mg _{Ir} cm ⁻²	1	0.4	0.1	MEA, PTL	High
Cathode Loading	mg _{Pt} cm ⁻²	0.5	0.1	0.025	MEA, GDL	Low
Membrane Thickness	μm	150	80	50	PEM	High
Cell Pressure	bar	<30	30	30	PEM	Low

7.1.1 Cost

System and stack CapEx greatly influence H₂ production costs. At the system level (which comprises the stack and balance of plant), increasing manufacturability capabilities in production volume and unit size (from kW to GW capacities) will be the primary driver for cost reductions. Other drivers include improving equipment utilization, utilizing roll-to-roll assembly, and increasing automation. At the stack level, component costs dictate, particularly for the MEA, PTL, and BPs. While increasing manufacturing volume will reduce costs, material costs will begin to dominate at higher volumes (**Figure 7a**). As the MEA can comprise nearly half of the stack cost at these scales, PGM loadings must be reduced while maintaining activity and durability. Novel base materials and coatings for BPs and PTLs, which are currently made of titanium and platinum, also need to be developed. Stainless steel is a promising base material for both.

Lastly, production costs will highly depend on electricity prices and capacity factors (**Figure 4**). Load-following for cheap renewable electricity will be critical. Operating at high capacity factors will reduce costs substantially, but periods of low load are inevitable. Thus, grid control algorithms and management strategies are required.

7.1.2 Durability

Commercial systems use high PGM loadings for MEAs, PTLs, and BPs to ensure adequate durability, leading to prohibitively high costs for large-scale deployment. Thus, developing cost-effective components that can reach lifetime targets will be a primary research focus. Optimizing the CL-PTL interface will be imperative to mitigate degradation, especially at low loadings. Incorporating an MPL on the PTL surface is a promising strategy. Also, membrane durability will become increasingly relevant as thinner membranes are implemented. Reinforcements, recombinant catalysts, and radical scavengers can be incorporated to combat any increased susceptibility to mechanical and chemical damage. Lastly, cell components are currently optimized for constant load operation, but PEMWEs will likely operate dynamically for load following. Thus, degradation under such conditions must be thoroughly considered during durability testing.

Standardized accelerated stress tests are required to ensure objective comparison between datasets and to assess durability under industrially relevant conditions (i.e., load following). While no such tests are currently established, ASTs for each PEMWE cell component have been proposed in

Table 4.

7.1.3 Efficiency

Achieving targets for stack efficiency (80% LHV) and nominal operating conditions (5A cm⁻² @ 1.7V) will rely on improvements in the anode CL, PTL, and membrane. Developing highly active low-loading anode CLs will require creating homogenous, electrically connected networks. Utilizing TiO₂ supports can help, but the challenge of forming percolating iridium oxide networks needs to be overcome. Optimizing PTL properties—such as porosity and pore size—can reduce mass transport losses and increase catalyst utilization. Incorporating an MPL can also increase utilization by enhancing CL-PTL interfacial contact. Lastly, reductions in membrane thickness will be imperative due to decreased ohmic overpotentials, but care must be taken to limit degradation and gas crossover.

7.2 Manufacturability

Given the current deployment of PEMWEs, considerations for large-scale manufacturing have largely been overlooked in the literature. Thus, issues may arise when standard lab-scale fabrication techniques cannot be employed at high throughput. For example, most of the high-performing low-loading CLs in the literature were created using spray coating,^{137,175,481} which is less suitable for roll-to-roll manufacturing than methods involving sweeping a tool over a substrate (e.g., slot-die coating, gravure coating, and screen printing). Furthermore, since optimal ink composition varies between methods, transferring knowledge acquired from one method to another can be ineffective. The authors recommend that more focus be put on fabrication techniques that are more suitable for large-scale production. In addition, the PTL fabrication process must also be improved for reliable high-throughput manufacturing. In-line quality controls will be necessary to detect surface defects, impurities, edge warps, and inhomogeneities. Without stringent efforts to guarantee CL and PTL quality, stack-level failures can occur and impart significant setbacks to large-scale operations.

7.3 Policies

The number of countries implementing green hydrogen policies is growing rapidly, with ~50 countries to date. This trajectory must be continued to meet deployment targets. However, despite the ambitious policies presented, a vast discrepancy exists between active projects and projects in the planning stages. Projects up to final investment decision (FID) constitute only 7% of total announced projects, accounting for a mere 0.7 GW of PEMWE deployment. Given that 220 GW are required by 2030 to reach net zero emissions, policymakers must substantially accelerate green hydrogen projects.

8 Conclusion

The vast global participation for PEMWEs provides much confidence for the future. With concerted efforts in every developmental facet, the path to GW-scale deployment can be realized. Fundamental research for improving activity and durability while limiting costs is necessary. Large-scale manufacturability must be considered in conjunction with any developments. As technological readiness levels increase, a mature set of codes and standards must be established to ensure safe operation. Lastly, global green hydrogen policies must be monitored to ensure that set targets are reached. This review has captured the current state of PEMWEs and provided an overarching road map for gigawatt-scale deployment.

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