

Electrolyzer and Fuel Cell Recycling for a Circular Hydrogen Economy

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Electrolyzers and fuel cells will be crucial for achieving global clean hydrogen and industrial decarbonization goals. However, the nascent clean hydrogen sector faces uncertainties around material supply chains and technology end-of-life management. This work aims to guide the transition to a circular hydrogen economy by using process modeling, techno-economic analysis, and life cycle assessment to evaluate the material cost, energy use, greenhouse gas (GHG) emissions, toxicity, and water use of five potential recycling strategies for proton exchange membrane electrolyzers (PEMWE) and fuel cells (PEMFC). Hydrometallurgy, acid dissolution, and electrochemical dissolution are shown to offer 2–7 times improvement across all assessed metrics relative to the manufacturing of PEMWE and PEMFC from raw materials. Recycling can also lower the raw material demand, material cost, energy use, and GHG emissions associated with PEMWE and PEMFC deployment in the United States in 2050 by 23%, 19%, 21%, and 16%, respectively. This study provides key insights into the costs, benefits, and complexities of recycling strategies for PEMWE and PEMFC, aiding the development of a circular economy that is synergistic with clean hydrogen deployment.

grow to 6000 and 900 GW, respectively, by 2050.^[2] Electrolyzers and fuel cells have an expected lifetime of 10 years due to catalyst degradation^[4,5] and rely on critical and imported materials such as platinum (Pt), iridium (Ir), and graphite.^[2,6] Careful end-of-life (EoL) management could help minimize supply chain risks, lower the environmental impacts associated with electrolyzer and fuel cell production and disposal, and enable affordable and clean energy while maintaining responsible consumption and production according to Sustainable Development Goals 7 and 12.^[7]

Electrolyzer and fuel cell EoL will likely include recycling, especially when reuse is not viable due to changes in technology composition and performance. While circular economy strategies are being implemented for other renewable energy technologies such as photovoltaics,^[8–10] wind turbines,^[11] and lithium-ion batteries,^[10,12,13] recycling of

1. Introduction

Clean hydrogen (H₂) produced electrochemically from water and renewable electricity is expected to play a pivotal role in the decarbonization of the transportation, heating, chemical, and iron and steel sectors.^[1] To meet a clean H₂ demand of 100 million metric tons per year (million t y⁻¹), the United States (U.S.) will need to increase installed electrolyzer capacity from 0.17 gigawatts (GW) in 2020 to 1000 GW in 2050. Stationary fuel cell capacity for the conversion of clean H₂ into electricity will need to increase from 0.5 to 50 GW within the same time period.^[2,3] Globally, cumulative electrolyzer and fuel cell capacities have the potential to

electrolyzers and fuel cells is limited due to the small volumes of these low-maturity technologies and the complexity of their composition.^[14] For example, current proton exchange membrane water electrolyzers (PEMWE) typically contain a membrane electrode assembly (MEA) comprising a perfluorinated sulfonic acid (PFSA) polymer coated in a Pt catalyst for the H₂ evolution reaction (cathode) and an Ir catalyst for the oxygen evolution reaction (anode). The MEA is sandwiched between a titanium gas diffusion layer (GDL) on the anode side and a carbon paper GDL on the cathode side, followed by titanium bipolar plates to conduct electrical current, forming a cell. Multiple cells are combined in a stack and encased by stainless steel end plates (Figure 1). Proton exchange membrane fuel cells (PEMFC) similarly have a PFSA and Pt MEA, carbon paper GDLs, graphite bipolar plates, and stainless steel end plates, but lack Ir on the anode (Figure 1). PEMWE and PEMFC components are often produced by different suppliers, complicating the EoL reverse supply chain.^[14]

Recycling strategies for PEMWE and PEMFC vary from mature (hydrometallurgy and pyrohydrometallurgy) to early stage (electrochemical, acid, or solvent dissolution) (Figure 1). The most established recycling technologies focus on recovering valuable precious metals and are already applied at an industrial scale for platinum group metal (PGM) extraction from vehicle catalytic converters.^[15] Hydrometallurgy leaches PGMs from the MEA

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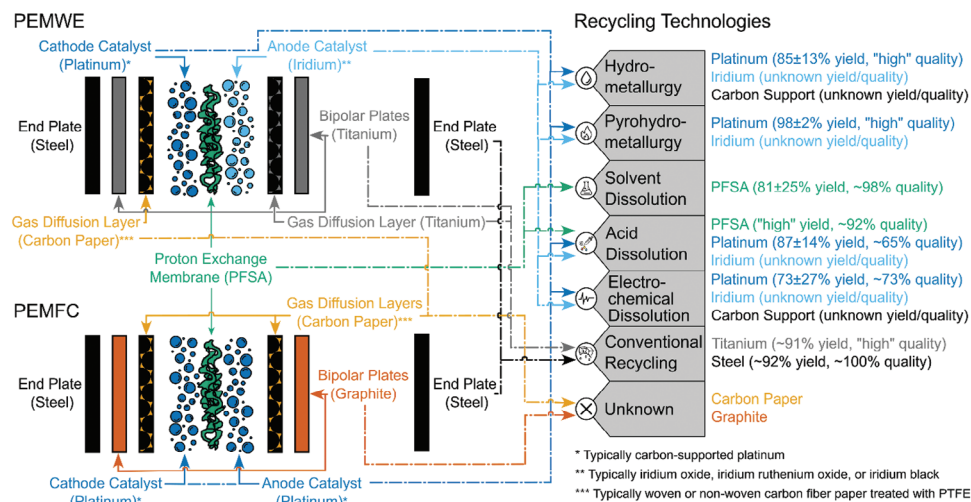


Figure 1. Breakdown of the material components of proton exchange membrane water electrolyzers and fuel cells (PEMWE and PEMFC) and their applicable recycling pathways. Data on the yields and qualities of recycled materials were sourced from the literature and are available in Table S1 (Supporting Information).

using an acid (HCl) and oxidant (H₂O₂ or HNO₃) and then recovers those metals through a series of extraction and separation steps involving organic solvents or ion exchange membranes.^[14] The yield of high-quality Pt recovered from PEMFC by hydrometallurgy has been reported to be 64–99% (Figure 1; Table S1, Supporting Information).^[16–19] Pyrohydrometallurgy first incinerates the PFSA and then proceeds with hydrometallurgy.^[14] This recycling technology has PGM yields of 96–100% (Figure 1; Table S1, Supporting Information) and eliminates the need for mechanical pretreatment, but releases fluorocarbon gases that can contribute to ozone depletion, global warming, and human health conditions.^[20] Electrochemical dissolution is a variation of hydrometallurgy in which PGMs are leached from the MEA using applied potentials and an acid (HCl).^[14] This early-stage technology uses milder conditions than hydrometallurgy but also has lower yields (25–95%) of lower-quality Pt (Figure 1; Table S1, Supporting Information).^[15,21,22] Definitions of Pt quality vary and may be linked to purity, particle size, or particle morphology, which will affect catalytic performance. Several emerging recycling techniques also focus on PFSA recovery, reducing fluorine emissions to the environment. Acid dissolution liquifies PFSA and PGMs in concentrated sulfuric acid (H₂SO₄) at elevated temperatures and separates the components through a series of extraction steps.^[14] Yields from PEMFC are similar to those of hydrometallurgy (65–100% for Pt), but the harsh conditions of acid dissolution can reduce the quality of the recovered catalysts and membrane (Figure 1; Table S1, Supporting Information).^[23] Solvent dissolution uses an alcohol-water mixture at elevated temperature to delaminate PFSA from the other MEA components,^[14] with high-quality PFSA yields of 63–99% (Figure 1; Table S1, Supporting Information).^[24–26] All technologies have been reported solely for PEMFC, which raises uncertainty around Ir recovery processes and yields for PEMWE. Bi-leaching – in which microorganisms produce chemicals that can leach metals from a solid matrix – has also been reported for PGM recovery from catalytic converters but not from PEMFC,^[27] and therefore is not studied in this work. Furthermore, data is

lacking on the recycling of other PEMWE and PEMFC components, including titanium, carbon paper, and graphite.

Here, we present a scoping analysis exploring the performance of PEMWE and PEMFC recycling by pyrohydrometallurgy, hydrometallurgy, acid dissolution, electrochemical dissolution, and solvent dissolution, in comparison to PEMWE and PEMFC manufacturing from raw materials. We develop process models for these recycling options and assess their cost, energy use, greenhouse gas (GHG) emissions, human toxicity, and water use by techno-economic analysis (TEA) and life cycle assessment (LCA). TEA and LCA are commonly used systematic methods for evaluating the economic and environmental viability, respectively, of a given process.^[28] We then leverage these results to estimate how the raw material demands, cost, energy use, and GHG emissions associated with PEMWE and PEMFC deployment in the U.S. from 2020 to 2060 could be reduced through recycling. This work quantitatively characterizes the challenges and opportunities facing PEMWE and PEMFC recycling, guiding decision-making toward a circular hydrogen economy that is economically viable and environmentally beneficial.

2. Results and Discussion

2.1. Comparison of Recycling Technologies

To estimate the costs and environmental impacts of PEMWE and PEMFC recycling strategies, we first built process models in Microsoft Excel based on data from the literature.^[19,21,23,24,29,30] For details, see Experimental Section and the process flow diagrams in Figures S1–S5 (Supporting Information). While these models reflect as much current published work as possible for the selected recycling technologies, they cannot encompass all potential process configurations that might result in TEA and LCA metrics that differ from those reported here. The models only considered the MEA portion of PEMWE or PEMFC; impacts and costs associated with dismantling and disposing of other stack components were not included. Furthermore, any additional

processing required to convert recycled Pt and Ir metals and PFSA solution into forms usable in PEMWE and PEMFC was not included; these steps were also excluded from the costs and impacts of raw Pt, Ir, and PFSA. Because all reported recycling technologies were for PEMFC (Pt only), we assumed that Ir yields from PEMWE would be equal to those of Pt and modeled the separation of Pt and Ir based on PGM solvent extraction processes.^[31] Pt and Ir have been recovered at similar yields to one another from hydrometallurgy of model PGM solutions and secondary PGM scrap.^[31,32] The material and energy requirements and capital costs from the process models (Tables S2–S6, Supporting Information) were used for TEA with a discounted cash flow analysis approach to estimate the minimum selling price (MSP, in U.S. dollars or USD) of the recycled materials. As defined here, MSP is the minimum acceptable selling price for recovered PEMWE or PEMFC materials that would enable a recycler to recoup their capital and operating costs (Table S7, Supporting Information). The process inventories were then used for LCA to estimate the energy use (gigajoules or GJ), GHG emissions (t carbon dioxide equivalent or t CO₂ eq), toxicity (t 1,4-dichlorobenzene equivalent or t 1,4-DCB eq), and water use (cubic meters of water or m³) of the recycled materials. Monte Carlo analysis was used to estimate the standard deviation of each reported metric.

For consistent comparison, results are reported for 1 megawatt (MW) of PEMWE or PEMFC MEA produced from 1 MW of MEA that enters a recycling process. Materials that are lost during recycling must therefore be replaced by raw materials. Market prices for these supplementary raw materials were sourced from industry databases and are the averages of 2017–2021 pricing (Table S8, Supporting Information).^[33,34] The combination of recycled material MSP and raw material market price is referred to as “Material Cost” and does not include factors such as manufacturing costs, manufacturer markup, or other market drivers that would determine final selling prices.^[35] The energy use, GHG emissions, toxicity, and water use associated with supplementary raw materials were obtained from the Material Flows through Industry (MFI) Tool orecoinvent databases (Table S8, Supporting Information).^[36,37]

Figure 2 shows the TEA and LCA results for PEMWE and PEMFC recycling (Table S9, Supporting Information). The lowest raw material demand (i.e., the highest recycled material yield) is achieved with acid or solvent dissolution due to their recovery of PFSA, which accounts for 85 wt.% of PEMWE MEA and 99 wt.% of PEMFC MEA (Table 1). For PEMWE (Figure 2A–F; Table S9, Supporting Information), pyrohydrometallurgy has the lowest material cost, energy use, toxicity, and water use because the high PGM yields (98% assumed here) reduce the quantities of raw Pt and Ir that are required. However, pyrohydrometallurgy also emits significant GHGs because the PFSA is assumed to be combusted into fluorocarbon gases with high global warming potentials,^[20,38] resulting in GHG emissions 44% higher than those of as-manufactured PEMWE. If pyrohydrometallurgy were to emit fluorocarbons with no global warming potential, such as carbonyl fluoride (COF₂), the GHG emissions of PEMWE recycling would decrease to 0.58 t CO₂ eq MW⁻¹, although toxicity would increase as COF₂ can react with water to form highly corrosive hydrofluoric acid.^[39,40] Hydrometallurgy and acid dissolution have similar material costs, energy

use, GHG emissions, toxicity, and water use at 33–34%, 24–28%, 21–25%, 13–15%, and 35–61% of the corresponding metrics for as-manufactured PEMWE. Previous LCA studies have similarly shown that PEMFC recycling by hydrometallurgy emits 1.5–6 times fewer GHG emissions and has 3 times lower toxicity than raw Pt.^[18,29,41] Approximately 40–70% of the material costs and environmental impacts of hydrometallurgy and acid dissolution originate in the raw Ir, Pt, and PFSA required to supplement the recycled materials; the remaining 30–60% is associated with the recycling processes themselves. Electrochemical dissolution has slightly poorer performance than the other recycling technologies across all metrics due to its lower PGM yields (73% vs 85% for hydrometallurgy and 87% for acid dissolution) and correspondingly higher raw PGM requirements, but its material cost and environmental impacts are still 28–45% those of as-manufactured PEMWE. Solvent dissolution has similar material costs and impacts to as-manufactured PEMWE because it focuses on PFSA recovery and raw PFSA is less expensive and impactful across all assessed metrics than raw PGMs (Table S8, Supporting Information).

While the results for PEMFC (Figure 2G–L; Table S9, Supporting Information) display similar trends to those discussed for PEMWE, two notable exceptions are that the energy use and GHG emissions of pyrohydrometallurgy increase substantially, and that acid dissolution becomes more competitive than hydrometallurgy across cost, energy use, and GHG emissions. These changes are due to the high PFSA to PGM ratio in PEMFC (168:1) in comparison to PEMWE (9:1, Table 1). As PFSA content increases, its recovery becomes more important and energy use and GHG emissions related to its incineration become more problematic. These results suggest that the optimal recycling technologies for PEMWE and PEMFC may be different, potentially complicating a future circular H₂ economy.

For both PEMWE (Figure S6, Supporting Information) and PEMFC (Figure S7, Supporting Information), 73–91% of the recycling technologies’ MSP originates in the salvage fee for the EoL stack, which was assumed to be 15% of the original stack material value. Here, the salvage fee refers to the residual value of an EoL PEMWE or PEMFC stack that is being decommissioned^[42]; this fee would be paid by a recycling facility to the stack operator. If the salvage fee is lower (5% of the original value), the material cost would be reduced by 24–55%, depending on the recycling process (Figure S8 and Table S10, Supporting Information). However, even if the salvage fee is 25% of the original stack value, pyrohydrometallurgy, hydrometallurgy, acid dissolution, and electrochemical dissolution all retain material costs 44–68% lower than PEMWE manufacturing from raw materials (Figure S8 and Table S10, Supporting Information). A salvage fee of up to 65% could be tolerated by the most expensive of these recycling technologies – electrochemical dissolution – without increasing material costs beyond as-manufactured PEMWE. The ability of recycling technologies to tolerate higher salvage fees could help decrease the leveled cost of energy from PEMWE or PEMFC and enable disassemblers to enter a circular economy market that might otherwise be considered unprofitable.^[43] Improving PGM yields to 99% could also reduce PEMWE and PEMFC recycling material costs by 10–58% and 6–47%, respectively, depending on the recycling process (Figures S8 and S9, Tables S10 and S11, Supporting Information). Capital expenses

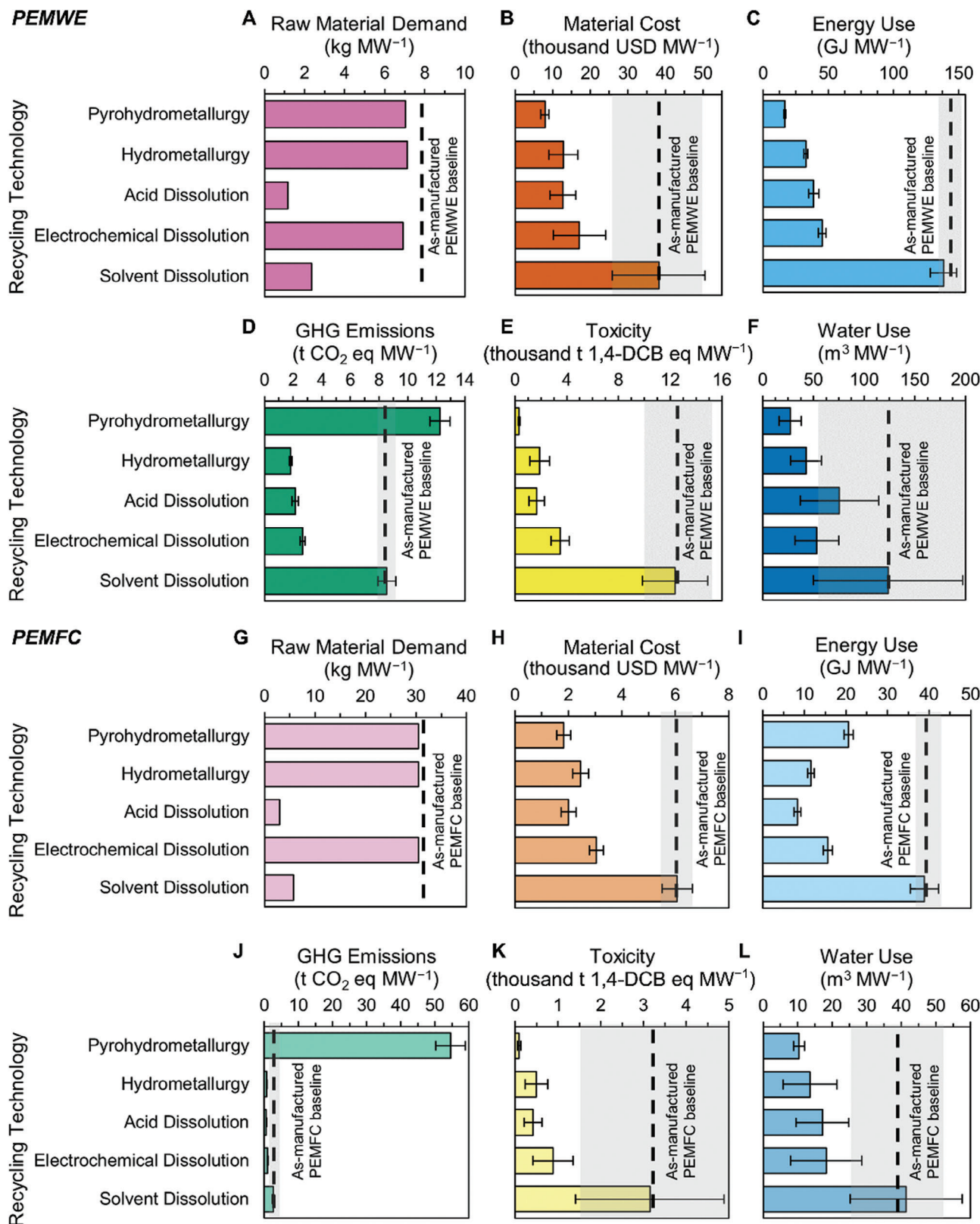


Figure 2. Comparison of the performance of PEMWE recycling strategies by A) raw material demand, B) material cost, C) energy use, D) GHG emissions, E) toxicity, and F) water use. Comparison of the performance of PEMFC recycling strategies by G) raw material demand, H) material cost, I) energy use, J) GHG emissions, K) toxicity, and L) water use. Results are reported on a per MW of output PEMWE or PEMFC basis. The gray rectangles indicate the standard deviations of the as-manufactured PEMWE or PEMFC baselines on a per MW basis. Raw data are available in Table S9 (Supporting Information).

Table 1. Material requirements assumed in this work for 1 MW of PEMWE or PEMFC. Data were sourced from the Department of Energy's Water Electrolyzers and Fuel Cells Supply Chain Deep Dive Assessment.^[2]

Material	Role	Quantity	Unit	Associated assumptions
PEMWE	Functional unit	1	MW	Cell area = 0.1225 m ² . Catalytic cell area = 0.0877 m ² . Stack size = 300 cells. Stack power = 1.0 MW
Iridium oxide	Anode	0.5262	kg	Loading = 2 mg cm ⁻²
of which Ir	Anode	0.4512	kg	
Carbon supported Pt	Cathode	0.6579	kg	Composition = 40%/60% Pt/C
of which Pt	Cathode	0.2632	kg	Loading = 1 mg cm ⁻²
of which carbon black	Cathode	0.3947	kg	
Titanium	Bipolar plate	248.4	kg	Thickness = 0.15 cm
Carbon paper	Cathode GDL	1.842	kg	Thickness = 215 μm. Density = 70 g m ⁻²
Titanium	Anode GDL	28.99	kg	Thickness = 0.025 cm
Stainless steel	End plates	20.95	kg	Thickness = 1 cm
PFSA	Proton exchange membrane	6.6174	kg	Thickness = 127 μm. Density = 1.98 g cm ⁻³
PEMFC	Functional unit	1	MW	Cell area = 0.1225 m ² . Catalytic cell area = 0.0877 m ² . Stack size = 300 cells. Stack power = 0.217 MW
Carbon supported Pt	Anode	0.1515	kg	Composition = 40%/60% Pt/C
of which Pt	Anode	0.0606	kg	Loading = 0.05 mg cm ⁻²
of which carbon black	Anode	0.0909	kg	
Carbon supported Pt	Cathode	0.3030	kg	Composition = 40%/60% Pt/C
of which Pt	Cathode	0.1212	kg	Loading = 0.1 mg cm ⁻²
of which carbon black	Cathode	0.1818	kg	
Graphite	Bipolar plate	574.0	kg	Thickness = 0.15 cm
Carbon paper	Cathode GDL	8.485	kg	Thickness = 215 μm. Density = 70 g m ⁻²
Carbon paper	Anode GDL	8.485	kg	Thickness = 215 μm. Density = 70 g m ⁻²
Stainless steel	End plates	20.95	kg	Thickness = 1 cm
PFSA	Proton exchange membrane	30.48	kg	Thickness = 127 μm. Density = 1.98 g cm ⁻³

account for 2–16% of recycled material MSP (Figures S6 and S7, Supporting Information). Pyrohydrometallurgy, which requires an expensive incinerator, and acid dissolution, which requires reactors compatible with concentrated H₂SO₄ and crystallizers for purification of the PFSA solution, are particularly capital-intensive recycling technologies.

Opportunities for improving the environmental metrics of the assessed recycling technologies vary. All pathways would benefit from increasing PGM yields to 99%, which could reduce the GHG emissions of PEMWE and PEMFC recycling by 49–81% and 45–58%, respectively (Figures S8 and S9, Tables S10 and S11, Supporting Information). Pyrohydrometallurgy impacts are further linked to natural gas use for PFSA incineration and to the emission of fluorocarbon gases (Figures S6 and S7, Supporting Information). Both factors could be challenging to mitigate, although potential solutions could include renewable heat sources (e.g., biogas) and flue gas scrubbing. Energy use, GHG emissions, toxicity, and water use of hydrometallurgy are dominated by the use of HCl (33–59% of impacts) and H₂O₂ (5–50%) in the leaching step, and by the use of NaOH (10–25%) and organic solvents (5–25%) for the Pt and Ir separation steps when applicable (Figures S6 and S7, Supporting Information). Similar trends are observed for electrochemical dissolution (Figures S6 and S7, Supporting Information). HCl and H₂O₂ consumption

could be reduced through wastewater recycling or less acidic leaching conditions, while NaOH and organic solvent use could be minimized by higher solvent recovery rates or by separating the PGMs by ion exchange membranes rather than solvent processes. Halving HCl concentration from 1 to 0.5 M or improving solvent recovery from 95% to 99% could reduce the GHG emissions of PEMWE hydrometallurgy by 2.5% and 2.6%, respectively (Figure S8 and Table S10, Supporting Information). Previous studies have also suggested that ion exchange membranes could reduce the GHG emissions of PEMFC hydrometallurgy by up to 15% relative to solvent separation, but that yield improvements for the membrane process are still necessary.^[18,29] The primary contributors to the environmental impacts of acid dissolution include steam for heating (2–65% of impacts), H₂SO₄ for the dissolution step (4–33%), and NaOH for neutralization (30–50%) (Figures S6 and S7, Supporting Information). Less extreme reaction conditions could be beneficial, such as halving H₂SO₄ concentration or lowering reaction time from 72 to 24 h for reductions in GHG emissions of 16% or 1.8%, respectively (Figure S8 and Table S10, Supporting Information). The energy use, GHG emissions, toxicity, and water use of solvent dissolution are dominated by steam for heating (3–60% of impacts) and butanol for the dissolution step (37–93%) (Figures S6 and S7, Supporting Information). GHG emissions could be reduced by 0.6% and 0.2%

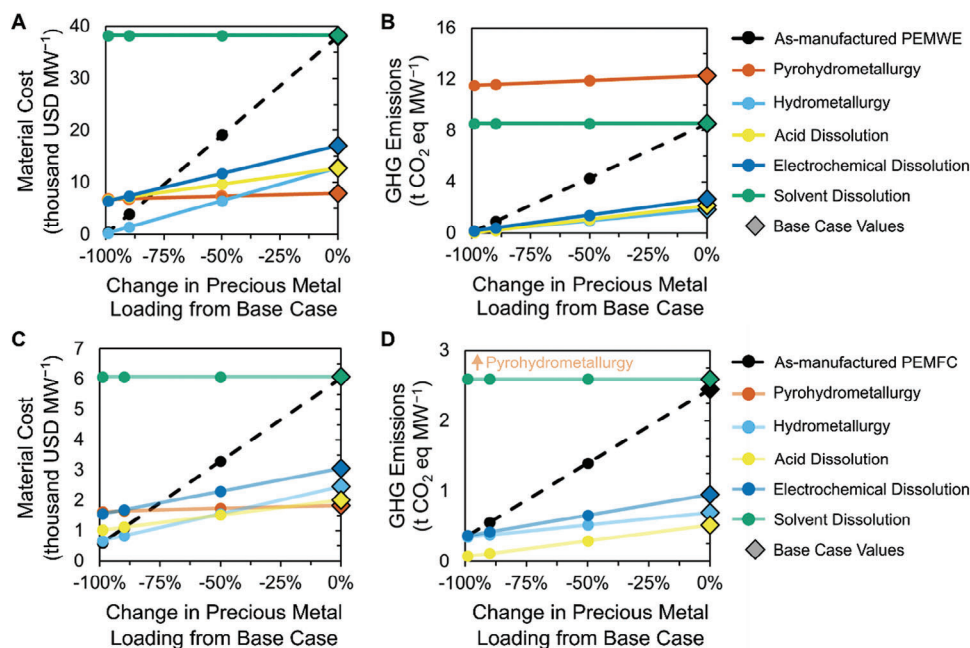


Figure 3. Sensitivity analysis of PGM loading variation from base case values (2 mg Ir cm^{-2} PEMWE anode, 1 mg Pt cm^{-2} PEMWE cathode, $0.05 \text{ mg Pt cm}^{-2}$ PEMFC anode, and $0.1 \text{ mg Pt cm}^{-2}$ PEMFC cathode; set at 0% in the figures). Results for PEMWE recycling (A) material cost and (B) GHG emissions. Results for PEMFC recycling (C) material cost and (D) GHG emissions. Raw data are available in Tables S10 and S11 (Supporting Information).

by increasing butanol recovery or decreasing reaction temperature from 100 to 50 °C, respectively (Figure S8 and Table S10, Supporting Information). Importantly, these various process improvements for the recycling technologies should be optimized to prevent yield losses, as yield is the most influential parameter on both material costs and environmental impacts (Figures S8 and S9, Supporting Information).

Future PEMWE and PEMFC technological changes may also affect the feasibility of recycling strategies. A major priority of PEMWE and PEMFC researchers is to reduce catalyst loading to minimize costs.^[44] While a lower cost could result in lower salvage fees, thereby reducing the cost of recycling, it could also limit the competitive advantage of recycling. For example, at PGM catalyst loadings one-tenth of our assumed values, all recycling technologies except for hydrometallurgy become more expensive than manufacturing PEMWE or PEMFC from raw materials (Figure 3). Similarly, the gap in GHG emissions between raw manufacturing and recycling by hydrometallurgy, acid dissolution, or electrochemical dissolution narrows from 3.2–4.6 times for PEMWE and 2.6–4.7 times for PEMFC with current catalyst loading values to 2.3–4.1 times for PEMWE and 1.3–1.5 times for PEMFC with one-tenth of that loading rate (Figure 3). In other words, recycling offers fewer GHG emission savings when catalyst loading decreases. Optimization analysis may be necessary to find a suitable compromise between stack cost and recycling feasibility. The performance and longevity of PEMWE and PEMFC stacks will also be affected by PGM loading rates and by the use of highly engineered materials that may prove more challenging to recycle.^[45,46] Holistic consideration of the tradeoffs between PEMWE and PEMFC capital cost, performance, durability, and

recyclability at EoL could therefore help inform future design and operational strategies for these systems.

2.2. Future Effects of Recycling

Lastly, we explored the U.S. economy-wide implications of PEMWE and PEMFC recycling by leveraging data on estimated future PEMWE and PEMFC deployment^[2] to assess the raw material demand, material cost, energy use, and GHG emissions of a Business as Usual Scenario and a Recycling Scenario (Figure 4). The Business as Usual Scenario assumes that no recycling occurs, meaning that all requirements are met by raw materials. The Recycling Scenario applies solvent dissolution (PFSA recovery), hydrometallurgy (PGM and carbon support recovery), and conventional recycling (titanium and stainless steel recovery). The combination of solvent dissolution and hydrometallurgy was selected as it exhibited the most optimal combination of low raw material demand, material cost, energy use, GHG emissions, toxicity, and water use, and high recycled material quality (Figure S10 and Table S9, Supporting Information). The results are depicted for the total PEMWE or PEMFC stack, not just for the MEA. See Experimental Section for further details.

The largest annual material demand expected to arise from clean H₂ deployment on a mass basis is titanium for the PEMWE anode GDL and bipolar plates (Figure 4A). The Recycling Scenario could reduce overall material demand by up to 23% in 2050 and up to 86% in 2060 (Figure 4B). This Scenario could also lower reliance on raw Ir and Pt by 28% in 2050 and 98% in 2060; these materials are currently 100% and 79% imported into

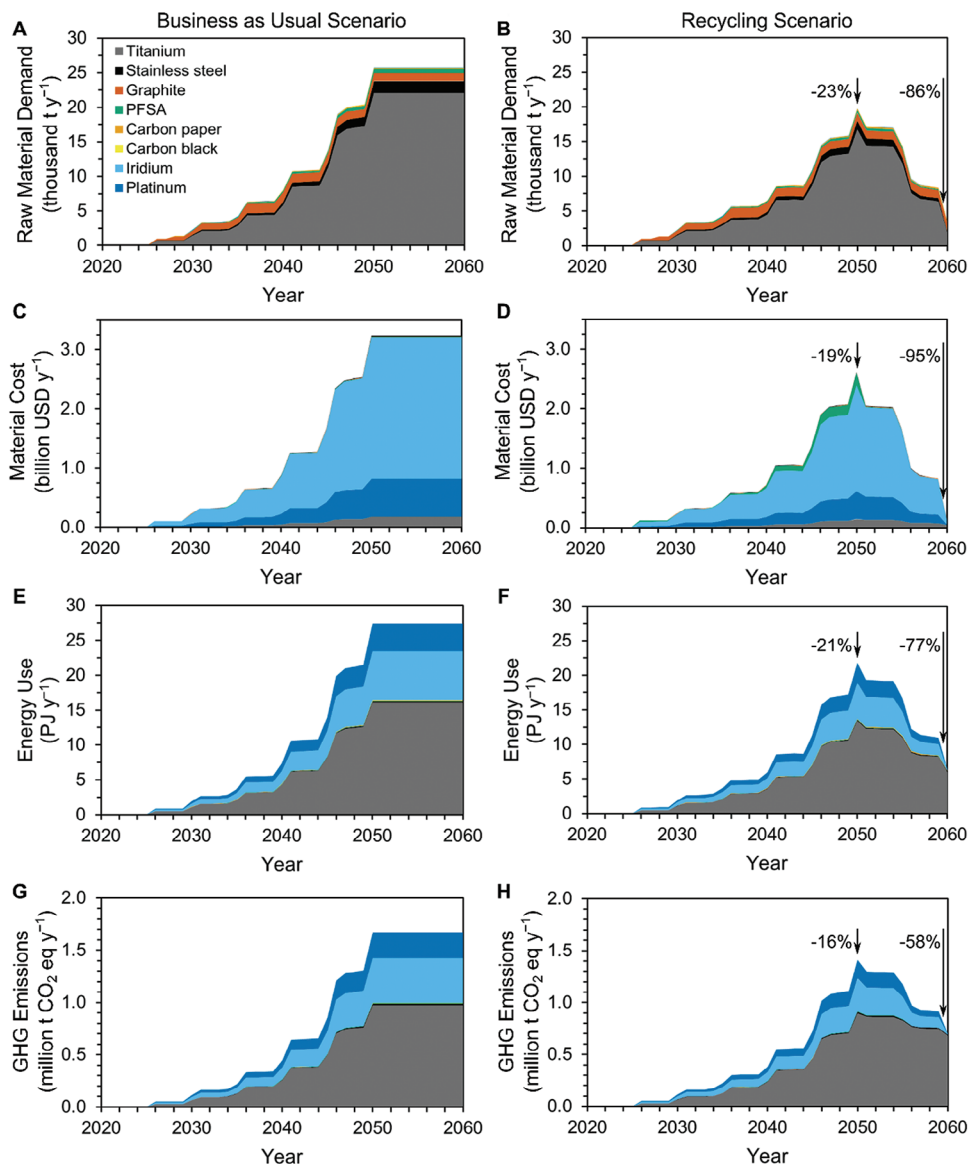


Figure 4. Annual A,B) raw material demand, C,D) material cost, E,F) energy use, and G,H) GHG emissions associated with U.S. PEMWE and PEMFC capacity without recycling (Business as Usual Scenario) or with recycling (Recycling Scenario). The Recycling Scenario includes solvent dissolution for PFSA recovery, hydrometallurgy for PGM and carbon support recovery, and conventional titanium and stainless steel recycling, and assumes 10-year PEMWE and PEMFC lifetimes. The percentages indicate reductions in each metric for the Recycling Scenario relative to the Business as Usual Scenario in 2050 and 2060.

the U.S., respectively.^[2] Material cost is dominated by Ir and Pt (Figure 4C). Recycling enables a 19% reduction in material cost by 2050 and a 95% reduction in material cost by 2060 (Figure 4D). Energy use (Figure 4E) and GHG emissions (Figure 4G) are dependent on the use of both PGMs and titanium. The Recycling Scenario could lower energy use by 21% in 2050 and 77% in 2060 (Figure 4F) and lower GHG emissions by 16% in 2050 and 58% in 2060 (Figure 4H). While the maximum GHG emissions associated with PEMWE and PEMFC manufacturing in the Business-as-Usual Scenario (1.67 million t CO₂ eq) represent less than 0.1% of total U.S. GHG emissions today (5588 million t CO₂ eq in 2021),^[47] recycling will nevertheless play a role in meeting U.S. net-zero targets.

Implementing the Recycling Scenario for a circular H₂ economy will require additional research and technology deployment. Several recycling strategies show preliminary potential over PEMWE and PEMFC manufacturing from raw materials, but further improvements could be achieved by increasing the material yields and qualities of these recycling processes while reducing energy and chemical use through less extreme reaction conditions, as well as exploring differences between recycling of PEMWE and PEMFC. While our analysis suggests that PFSA recycling by solvent dissolution may currently be more impactful and costly than raw PFSA manufacturing, closing the fluorine loop to minimize fluorine emissions into the environment or landfills is also a key consideration. Furthermore, there

is a lack of data on the recycling of other stack components such as graphite and carbon paper, as well as on potential mismatches between the high titanium grade required for electrolyzers and the lower titanium grade typically available from manufacturers and recyclers.^[48] From a stack engineering perspective, PEMWE and PEMFC could be better designed for disassembly (e.g., using bolts rather than sealants)^[49] and recycling (e.g., using the same type of polymer for both the membrane and catalyst adhesives, or exploring alternative GDL materials).^[50] Furthermore, automation for disassembly, which is being explored for lithium-ion battery recycling, could help reduce costs and worker safety impacts.^[51] From a deployment perspective, PEMWE and PEMFC suppliers would likely benefit from streamlining the EoL reverse supply chain and leveraging co-processing with other types of electronic waste, such as batteries or rare earth magnets. Lastly, further exploration of the environmental and social justice impacts of these recycling processes – as well as of recycling options for solid oxide electrolyzers and fuel cells – could help ensure that deployment brings benefits rather than disadvantages to the local communities in which they are sited.

3. Conclusion

Recycling electrolyzers and fuel cells can improve supply chain resiliency and minimize the environmental impacts and costs of a clean H₂ economy. We explored the feasibility of five recycling strategies for PEMWE and PEMFC through process modeling, TEA, and LCA. Hydrometallurgy, acid dissolution, and electrochemical dissolution all featured material costs, energy use, GHG emissions, toxicity, and water use 2–8 times lower than conventional PEMWE or PEMFC manufacturing. The remaining technologies exhibited important tradeoffs: pyrohydrometallurgy was shown to be a low-cost method for obtaining high yields of high-quality PGMs, but it emits copious GHGs due to PFSA incineration, while solvent dissolution is able to recover PFSA but at less competitive costs and higher environmental impacts than raw materials. Furthermore, the application of consecutive solvent dissolution and hydrometallurgy could reduce material costs and environmental impacts and secure a more resilient supply chain for clean H₂ deployment in the U.S. Key opportunities in the PEMWE and PEMFC recycling space were identified to include process yield and quality improvements, harsh chemical and energy use minimization, closing of the fluorine loop, PEMWE and PEMFC design for disassembly and recycling, and co-processing with other electronic waste sources. Overall, this work showcased the benefits and risks of PEMWE and PEMFC recycling strategies, helping to promote the development of a more sustainable, cost-effective, and circular economy for clean H₂.

4. Experimental Section

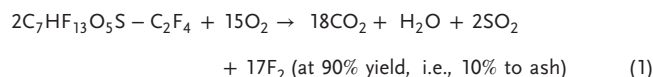
Scope: The functional unit of this work is the production of 1 MW of PEMWE or PEMFC from 1 MW of PEMWE or PEMFC that enters a recycling process. Any materials that are lost during the recycling process due to low yields must therefore be replaced by raw materials. The material requirements for 1 MW PEMWE or PEMFC are listed in Table 1.^[2] The input of all process models was assumed to be MEAs that have been removed from the remainder of the PEMWE or PEMFC. Dismantling of the

PEMWE or PEMFC was not included in the present models but would likely add costs and energy requirements to the overall recycling processes.^[52] Stainless steel and titanium components were assumed to be recyclable by conventional melt processing.^[48,53] Carbon paper and graphite components were assumed to be non-recyclable and sent to landfill.^[54,55] The metrics assessed in this study are listed in Table 2.

Process Models: Process models for each recycling technology were constructed in Microsoft Excel on a 1 t PGM y⁻¹ basis and can be made available upon request. Process flow diagrams for all processes are presented in Figures S1–S5 (Supporting Information).

The hydrometallurgy process was modeled based on Duclos et al. (Figure S1 and Table S2, Supporting Information).^[19,29] The MEA was first ground into small particles. The PGMs were then leached from the carbon support and PFSA membrane at 70 °C for 24 h in the presence of 1 m aq. HCl and 3 vol% H₂O₂ with stirring at a concentration of 0.002 kg metal L⁻¹ H₂O. The heat was assumed to be provided by low-pressure steam. The undissolved carbon support and membrane were filtered off and sent to waste treatment. For PEMWE, the Pt (in the form of chloroplatinic acid) was extracted from the aqueous phase into an organic phase (15 vol% Cyanex, 85 vol% toluene, at an aq:org volume ratio of 1:1).^[19,29] Ir was assumed to be in a reduced state due to the presence of H₂O₂ and must be re-oxidized by 1 m NaClO₃ to be extracted into an organic phase separate from that of Pt.^[31] The PGMs were stripped by 2 m NaOH into a clean aqueous phase (org:aq volume ratio of 1:1). The PGMs were then precipitated as ammonium salts in the presence of 0.025 m NH₄Cl and 1.5 m HCl,^[19,29] filtered from solution, and reduced by H₂ to form Pt or Ir metals suitable for catalytic applications.^[56] For PEMFC, the aqueous chloroplatinic acid phase was sent directly to the reduction step. The overall PGM yields and qualities were assumed to be 85% and 100%, respectively, based on the literature (Table S1, Supporting Information).

The pyrohydrometallurgy process was modeled based on Zhao et al. (Figure S2 and Table S3, Supporting Information).^[30] The MEA was first dried at 80 °C for 3 h and then heated at 600 °C for 6 h in the presence of air. The heat was assumed to be provided by fired heat. Combustion of PFSA was approximated using:



F₂ was assumed to be emitted as fluorocarbon gases,^[20] with unknown composition and an emission factor of 3236 kg CO₂ eq kg⁻¹ F₂ (averaged from the emission factors of 26 common (hydro)fluorocarbon gases).^[38] The PGM and ash mixture was sent to a hydrometallurgical leaching chamber for 12 h (see above). All subsequent extraction and reduction steps were kept consistent with those reported for hydrometallurgy. The overall PGM yields and qualities were assumed to be 98% and 100%, respectively, based on the literature (Table S1, Supporting Information).

The acid dissolution process was modeled based on Xu et al. (Figure S3 and Table S4, Supporting Information).^[23] The MEA was first cleaned in 0.5 m aq. H₂SO₄ for 2 h at 25 °C at a concentration of 0.02 kg metal L⁻¹ H₂O. The MEA was dried, ground into small particles, and dissolved in 18 m aq. H₂SO₄ at 150 °C for 72 h with stirring. The heat was assumed to be provided by low-pressure steam. CO₂ and SO₂ emissions from the reaction of H₂SO₄ with the carbon support were approximated using:



The PGMs were precipitated using NaOH (2 mol per mol H₂SO₄) and separated by centrifugation. The remaining neutralized aqueous phase was cooled to crystallize and remove a Na₂SO₄ co-product, leaving an aqueous phase containing 3–10 wt.% PFSA, which can be re-cast into a membrane. For PEMWE, the Ir and Pt sulfate salts were dissolved in 1 m aq. HCl at a concentration of 0.002 kg metal L⁻¹ H₂O and subsequently extracted and reduced as described for hydrometallurgy. For PEMFC, the Pt sulfate salt was directly reduced to a form suitable for catalysis. The overall PGM yields and qualities were assumed to be 87% and 65%, respectively, while

Table 2. List of assessed metrics and their definitions. For descriptions of TEA, MFI, and LCA, refer to the “Economic analysis” and “Environmental analysis” portions of the Experimental Section.

Metric	Definition	Method	Unit
Quality	Performance of the recycled material divided by that of the raw material, typically characterized by electrochemical active surface area for PGMs or voltage under an applied current for PFSA	Literature review	%
Yield	Quantity of material output from recycling divided by quantity of material input to recycling	Literature review	%
Raw Material Demand	Inverse of yield	Calculation	kg
MSP	Minimum acceptable price of a recycled material required to recoup operational and capital costs	TEA	USD
Market Price	Selling price of raw materials	Literature review	USD
Material Cost	Combination of MSP for recycled materials and market prices for raw materials	Calculation	USD
Energy Use	Supply chain energy used by a given material or process	MFI	MJ
GHG Emissions	Supply chain GHG emissions produced by a given material or process	MFI	kg CO ₂ eq
Toxicity	Risk increase of cancerous and non-cancerous disease incidence	LCA	kg 1,4-DCB eq ^a
Water Use	Increase in global water consumption	LCA	m ³

^a) 1,4-DCB = 1,4-dichlorobenzene.

those of the PFSA were assumed to be 90% and 92%, respectively, based on the literature (Table S1, Supporting Information).

The electrochemical dissolution process was modeled based on Lat-suzbaia et al. (Figure S4 and Table S5, Supporting Information).^[21] The MEA was first cleaned in 0.1 M aq. HClO₄ (1 mL H₂O cm⁻² MEA) by cycling from 0.05 to 1.4 V for 10 cycles at a rate of 50 mV s⁻¹ in an electrochemical cell under an inert atmosphere. The HClO₄ solution was replaced by a 0.1 M aq. HCl solution (1 mL H₂O cm⁻² MEA), and the electrochemical cell was purged with oxygen. The cell was cycled from 0.5 to 1.1 V for 1000 cycles at a rate of 50 mV s⁻¹ (≈3.33 h and 5.2E–5 kWh m⁻² MEA cycle⁻¹). During this process, the PGMs dissolved into the aqueous solution, and the carbon support delaminated from both the PFSA and PGMs. The PFSA was removed and sent to waste disposal, while the carbon support was recovered by filtration. The PGMs were subsequently extracted and reduced as described for hydrometallurgy. The overall PGM and carbon support yields and qualities were assumed to be 73% and 73%, respectively, based on the literature (Table S1, Supporting Information).

Finally, the solvent dissolution process was modeled based on a BASF patent (Figure S5 and Table S6, Supporting Information).^[24] The MEA was first ground into small particles and then the PFSA was delaminated from the PGMs and carbon support in a butanol-water mixture (25 vol% butanol, 75 vol% water, 125 L kg⁻¹ PFSA) at 100 °C for 0.5 h. The heat was assumed to be provided by low-pressure steam. The PGMs and carbon support were filtered from the solution, dried, and could be sold to a downstream recycling process such as hydrometallurgy. The PFSA was separated from the residual solution by ultrafiltration and dried. The PFSA yield and quality were assumed to be 81% and 98%, respectively, based on the literature (Table S1, Supporting Information).

Economic Analysis: Material and energy balances from the process models were used to estimate raw material consumption, utilities, other variable operating expenses, equipment sizing, and capital investment. Pricing of consumables in the U.S. was obtained from industry databases (2017–2021 average).^[33,34] A salvage fee of 15% of the original PEMWE or PEMFC MEA material value was used.^[42] Capital equipment costs were obtained from the literature and adjusted to the process scale using a scaling exponent of 0.7.^[57–59] A discounted cash flow approach with financial parameters listed in Table S7 (Supporting Information) and an assumed plant lifetime of 30 years was applied to estimate the MSPs of material obtained from each recycling process on a per kilogram product basis. The overall material cost for 1 MW of PEMWE or PEMFC combined the recycled material MSP and raw material market prices and did not include assembly costs, manufacturer markup, or other market drivers that would determine final selling prices.

Environmental Analysis: Material and energy balances from the process models were used to build life cycle inventories for each recycling process (Tables S2–S6 and S12, Supporting Information). These life cycle inventories were input into the Material Flows through Industry (MFI) tool, which computes GHG emissions and energy use for a U.S. context.^[36] Toxicity and water use were calculated with SimaPro LCA software, ecoinvent v3.3 background data (allocation, cutoff by classification – unit, U.S.-specific inventories when available, global inventories otherwise),^[37] and the ReCiPe Hierarchist midpoint method.^[60] A cutoff approach was used in which postconsumer PEMWE or PEMFC were assumed to be free of environmental burdens. The overall environmental impacts were determined by combining the GHG emissions, energy use, toxicity, and water use of the recycled materials with those of any raw materials required to meet the requirements for 1 MW of PEMWE or PEMFC.

Uncertainty Analysis: Uncertainty was estimated using a semi-quantitative pedigree approach.^[61] Each item in the life cycle inventory was assumed to vary between ±20% according to reliability, completeness, temporal correlation, geographical correlation, and further technological correlation scores.^[61] The resulting ranges determined the low and high values of symmetric triangular distributions for each inventory item. Pricing uncertainty was assigned a triangular distribution based on average, low, and high annual costs between 2017 and 2021. Background data uncertainty for GHG emissions and energy use was assigned a triangular distribution based on average, lowest, and highest impact processes for a given material or energy source in MFI. For toxicity and water use, the uncertainty of background data was provided by lognormal distributions in ecoinvent. With these distributions, a Monte Carlo analysis was performed with 1000 iterations, giving mean and σ values. Error propagation was applied when necessary.

Sensitivity Analysis: Sensitivity analysis was conducted to determine how the alteration of key variables impacts the performance metrics of the recycling processes.

Future Projections: Data on future electrolyzer and fuel cell deployment and the corresponding material requirements were obtained from the Department of Energy’s Water Electrolyzers and Fuel Cells Supply Chain Deep Dive Assessment.^[2] In brief, this report assumes that the U.S. will produce 100 million t y⁻¹ of clean H₂ by 2050 in order to meet decarbonization goals and that PEMWE will supply 54% of this demand (the remainder is supplied by solid oxide and alkaline electrolyzers). All material demands for the Business as Usual Scenario from 2020 to 2050 were obtained from the raw data behind the Deep Dive Assessment and can be made available upon request. Clean H₂ demand and the corresponding material demand were assumed to remain constant after 2050. Material demands in the

Recycling Scenario were obtained by assuming a combination of solvent dissolution (81% PFSA recovery), hydrometallurgy (85% PGM recovery), conventional recycling (91% titanium recovery, 92% stainless steel recovery), and a PEMWE or PEMFC lifetime of 10 years (i.e., the material available for recycling in 2030 is the material that was originally used in 2020). The recycling yields of Pt and Ir were assumed to slowly increase over time by the learning-by-doing effect to 98% in 2050. To obtain material costs, GHG emissions, and energy use, the material demands from the Business as Usual Scenario or the Recycling Scenario were multiplied by the MSPs or market prices, GHG emissions, and energy use of the respective materials (Table S8, Supporting Information). Raw material data were sourced from the literature, recycled PGM, carbon black, and PFSA data were calculated from the combined solvent dissolution and hydrometallurgy process model, and recycled titanium and steel data were obtained from the literature.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available in the supplementary material of this article.

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electrolyzer, fuel cell, life cycle assessment, recycling, techno-economic analysis

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