

Lawrence Berkeley National Laboratory

LBL Publications

Title

Advances and prospects to achieve high-performing and durable proton-exchange-membrane unitized regenerative fuel cells

Permalink

<https://escholarship.org/uc/item/512076bx>

Authors

Wang, Shiyi

Weber, Adam Z

Peng, Xiong

Publication Date

2023-08-01

DOI

10.1016/j.coelec.2023.101340

Copyright Information

This work is made available under the terms of a Creative Commons Attribution-NonCommercial-NoDerivatives License, available at <https://creativecommons.org/licenses/by-nc-nd/4.0/>

Peer reviewed



ELSEVIER

Review Article

Advances and prospects to achieve high-performing and durable proton-exchange-membrane unitized regenerative fuel cells

Shiyi Wang, Adam Z. Weber and Xiong Peng

**Abstract**

Proton-exchange-membrane unitized regenerative fuel cell (PEM-URFC) is a promising energy storage and conversion device for large-scale and long-term applications. Previous research has primarily focused on materials development studies, with performance and durability not evaluated at relevant conditions. Such an approach becomes insufficient for making URFC technology commercially competitive. In this review, we highlight recent progress on high-performing PEM-URFCs, focusing on electrode engineering, key components, and operating approaches that take realistic operating conditions into consideration. Key components of a membrane electrode assembly (MEA), including catalyst layer, diffusion media, and membrane, which require different optimization strategies, are discussed in this review.

Addresses

Energy Storage and Distributed Resources Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA

Corresponding author: Peng, Xiong (xiongp@lbl.gov)

Current Opinion in Electrochemistry 2023, 40:101340

This review comes from a themed issue on **Electrochemical Materials and Engineering (2023)**

Edited by **Frederic Jaouen** and **Chang-Hyuck Choi**

For complete overview about the section, refer [Electrochemical Materials and Engineering \(2023\)](#)

Available online 17 June 2023

<https://doi.org/10.1016/j.coelec.2023.101340>

2451-9103/© 2023 The Author(s). Published by Elsevier B.V. This is an open access article under the CC BY license (<http://creativecommons.org/licenses/by/4.0/>).

Keywords

Unitized regenerative fuel cell, Bifunctional electrode, Proton exchange membrane, Porous transport layer, Durability.

Introduction

The proton-exchange-membrane regenerative fuel cell (PEM-RFC) is an energy conversion and storage device, which stores electrical energy in the form of H₂ and O₂ gas by splitting water through the oxygen evolution reaction (OER) and the hydrogen evolution reaction (HER) during charging, and regenerates electricity from the stored H₂ and O₂ gas by reforming

water through the oxygen reduction reaction (ORR) and the hydrogen oxidation reaction (HOR) during discharging. This process can be achieved by integrating two distinct compartments: a water electrolyzer (WE) unit for charging and a fuel cell (FC) unit for discharging. A compact design of RFC is the unitized regenerative fuel cell (URFC), which combines the discrete WE and FC into a single device and offers capital cost reduction.

The development of PEM-URFCs can be traced back to the early 1970s [1,2], for niche applications such as aerospace, aviation, or unmanned vehicle [3]. Nowadays, due to environmental concerns, a growing trend of utilizing renewable energy such as solar and wind to decarbonize various sectors has been observed worldwide. However, due to the intrinsic intermittency, it is extremely challenging to rely on very high shares of wind or solar to achieve deep decarbonization. The PEM-URFC offers a practical solution to managing these intermittent and variable energy sources owing to its high scale-up efficiency benefited from the decoupled energy storage capacity and rated power, long cycle life, low weight and volume, and significant advantages in specific energy [4]. However, PEM-URFC remains less commercially competitive compared to other energy storage solutions, limited by its low round-trip efficiency (RTE) and durability. The RTE is a widely used criterion to evaluate the performance of energy conversion and storage device, which is defined as the percentage of the amount of energy that can be recovered (discharging) relative to the energy stored (charging). Specifically for URFC, RTE can be calculated as the product of WE mode voltage efficiency and FC mode efficiency (efficiency loss from H₂ crossover is not considered, detailed calculation can refer to the supplementary material of Ref. [5]). Development of PEM-URFC technology has mainly focused on advancing the bifunctional performance of cell materials; however, most reported RTEs are less than 50% at low currents, which falls short of widespread commercialization criteria such as lithium-ion batteries, whose RTEs can reach >90%. In recent years, breakthroughs in PEM-URFC design are achieved by optimizations at the cell level rather than materials innovation, including electrode design, cell assembly, and operating strategies [5–7]. In this review, we will

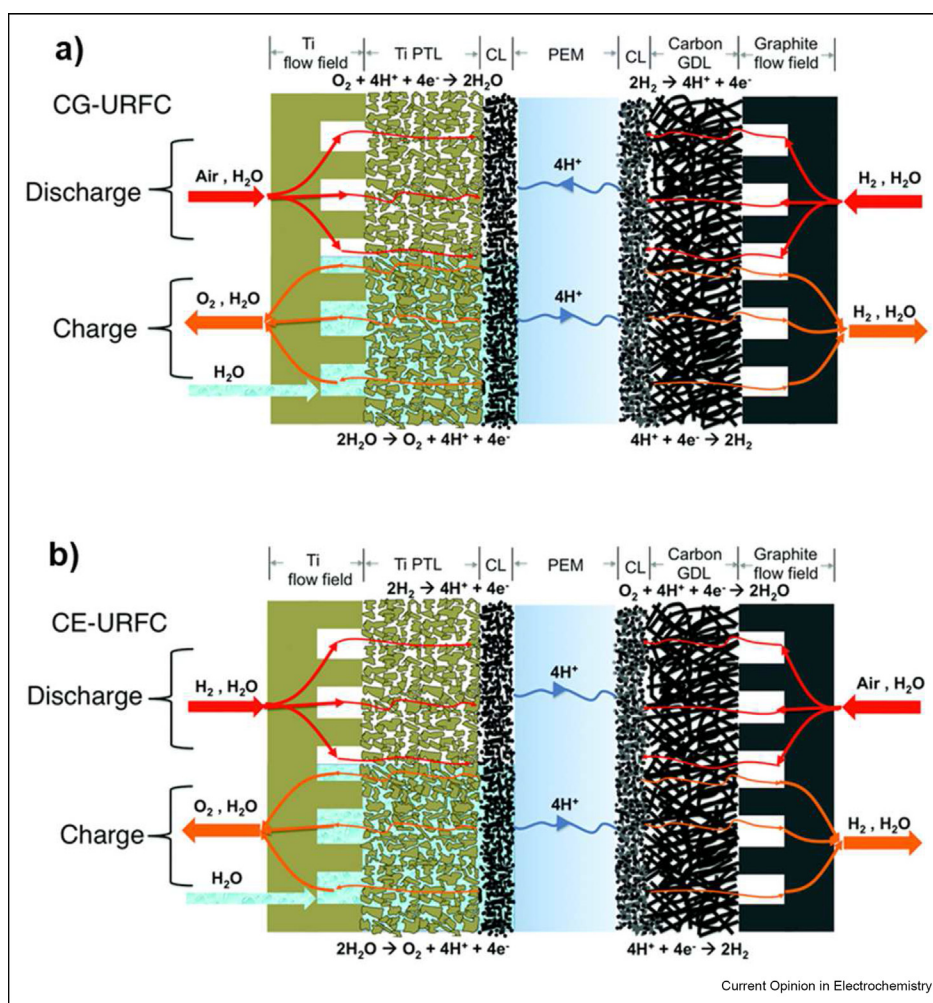
address the recent progress made in PEM-URFC technology through these approaches.

PEM-URFC components and operation modes

The membrane electrode assembly (MEA) in a PEM-URFC device is more similar to WE rather than FC. Unlike the carbon-based materials used in both electrodes in FC, materials such as Ti, Ir, Ru, and their oxides are required for OER electrode to avoid fast degradation during WE operation [8–10]. Conventionally, URFCs are often operated in constant-gas (CG) mode where ORR and OER share one electrode and HER and HOR share the other electrode (Figure 1a). Such operating mode leads to the disadvantage that the two performance-determinative reactions, ORR and OER, occur at the same side, which restricts performance and durability. An alternative operating mode is the constant-electrode (CE) where ORR and HER

share one electrode and OER and HOR share the other electrode (Figure 1b). Regmi and coworkers suggest that URFC operating in CE configuration significantly improves the performance compared to CG configuration, which also offsets the expense of N₂-purge required for CE operation, leading to a lower levelized cost of storage (LCOS) from a cost analysis assuming equivalent daily duty and lifetime [6]. Better CE configuration performances over CG are also reported by Peng et al. [5] and Ahyoun et al. [11]. CE mode operation allows ORR to occur at the carbon-based electrode with the preferred structure and thus reducing the mass transport resistance [5]. This operation strategy avoids major challenges in developing bifunctional oxygen electrodes that CG mode operation encounters. Potential drawbacks of CE operation includes safety concerns due to possible H₂ and O₂ mixing [6], and durability issues due to the broader operating potential window for each electrode.

Figure 1



Schematic illustration of PEM-URFC operated with (a) CG configuration and (b) CE configuration. Adapted with permission from Ref. [5].

However, the notable efficiency improvement achieved by simply switching from CG to CE operation makes it a promising research direction for PEM-URFC design. Regular reporting of PEM-URFC performance and durability in both CG and CE modes is something researchers should consider in this field.

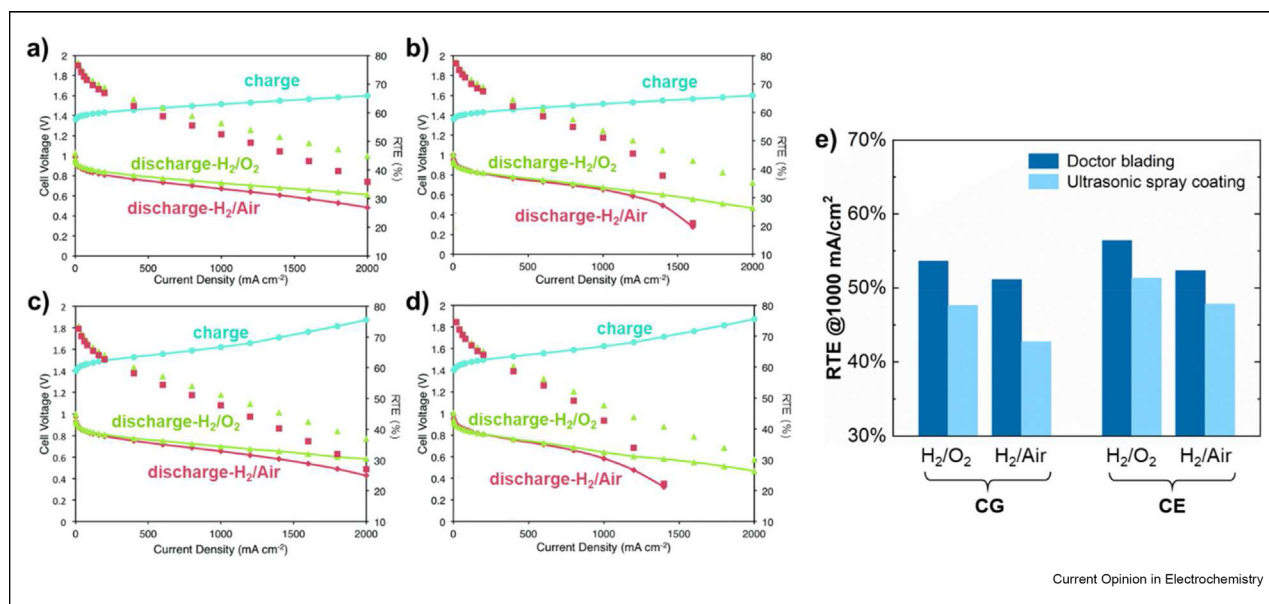
Bifunctional catalysts and bifunctional electrodes

Previous efforts for PEM-URFC have been mostly focused on bifunctional OER/ORR catalyst development [12–19]. To enable OER and ORR at the same electrode during CG operation, Pt, Ir, Ru, and their oxides are typically alloyed to improve the bifunctional kinetics and maintain stability. Recent reports include exploration of the catalyst composition [17], catalyst morphology control [13,18], alloy stoichiometric number [14–16], and catalyst support [12,19,20]. The kinetic properties of bifunctional catalysts are the main focus of improvement; for example, electrochemical performance is either evaluated by a rotating disk electrode (RDE) system or MEA at low current densities ($<0.5 \text{ A/cm}^2$) [21–25]. However, there have been very few catalyst studies that turn into significant advancements for PEM-URFCs.

The catalyst layer plays a vital role in mass transport, water management, and catalyst utilization besides

governing the kinetics, which affects URFC performance across a wide range of operating conditions [5,26]. Quite frankly, developing a bifunctional OER/ORR catalyst is not necessary as physically mixing two active OER and ORR to achieve the bifunctional catalyst layer is probably more efficient. Thus, optimizing a bifunctional electrode with performance evaluated at realistic operating conditions is more informative for guiding high-performance PEM-URFC design. Kúš and colleagues report RTEs of 31.8% at 1 A/cm^2 under CE operation using a sandwich-like thin-film TiC-supported Pt-Ir electrode synthesized via magnetron sputtering [27]. Peng *et al.* point out that rather than the intrinsic catalyst activity, the low RTEs are likely attributed to the severe mass transport resistance and low utilization of catalysts induced by fabrication-dependent catalyst layer structure, especially at low catalyst loading [5]. Two types of catalyst-coated membrane (CCM) are fabricated by mixing Pt black and Ir black using ultrasonic spray coating and doctor blading approaches with related ink recipes. The doctor blading method achieves enhanced cell performance, reaching RTEs of 56.4% and 52.3% under CE operation, 53.6% and 51.1% at 1 A/cm^2 under CG operation with H_2/O_2 and H_2/air feeds, respectively (Figure 2). These studies indicate that electrodes with high porosity and low tortuosity are the preferred design for PEM-URFCs. For PEM-URFC under CE mode, we believe that the design

Figure 2



Charging and discharging polarization curves under CE and CG mode with H_2/O_2 and H_2/Air feeds in PEM-URFCs using bifunctional electrodes with different fabrication methods. RTEs are illustrated for performance comparison. Pt-Ir electrode using doctor blading method with a) CE and b) CG configuration operation. Pt-Ir electrode using ultrasonic spray coating with c) CE and d) CG configuration operation. RTEs are shown as scattered points with corresponding color codes. e) Comparison of RTEs at 1000 mA/cm^2 extracted from a) to d) under different operating conditions. Electrode fabricated with the doctor blading method leads to higher RTEs compared to the ultrasonic spray coating method. Adapted with permission from Ref. [5,27]. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

principle of OER/HOR and ORR/HER bifunctional electrode can be conveniently learned from WE anode and FC cathode studies respectively, as the electrode for these two performance-determining reactions (OER and ORR) can be optimized separately and the hydrogen catalysis are typically less of a concern. The ideal catalyst layer should be optimized not only in kinetics, but also catalyst utilization and mass transport, and we encourage more research efforts focusing on bifunctional electrode design using CE operating mode. Besides, the durability of OER/HOR and ORR/HER catalysts or electrodes needs to be designed to accommodate a wider range of operating potential under CE mode.

Design of porous transport layers

The diffusion media is the core component that governs the transport of the two-phase flow during both WE (charging) and FC (discharging) operations. Titanium-based porous transport layers (Ti PTLs) are typically used in charging to avoid fast corrosion. The hydrophilic nature of PTLs ensures water supply during charging, however, makes it less favorable for water removal and gas transport during discharging especially under CG mode. Tuning the hydrophobicity of PTLs to balance charging and discharging can be critical for high RTEs. Relevant research includes using polytetrafluoroethylene (PTFE) coating to render the hydrophobicity of PTLs and exploring the optimal PTFE loading and fabrication method. Omrani and Shabani have reviewed related reports and indicated that the impacts of PTFE coating parameters can be arbitrary [10]. They suggest that optimal PTFE loading is dependent on operating conditions and coating processes [28]. Lim et al. note that although the addition of PTFE improved FC performance, it inevitably sacrifices WE performance. In their recent report, an amphiphilic Ti PTL is fabricated by introducing alternative hydrophobic and hydrophilic channels using a shadow mask patterning process with an ultra-thin polydimethylsiloxane brush as the hydrophobic surface modifier [11]. The PEM-URFC performance is improved by 4.3 times in FC mode (at 0.6 V) and 1.9 times in WE mode (at 1.8 V) compared to using a pristine PTL. Zhang et al. test PEM-URFC using a hybrid wettability carbon-based GDL with alternative hydrophobic and hydrophilic channels that integrated with the flow channel of a bipolar plate, which achieves better performance in both FC and WE operations compared to using an untreated GDL [29].

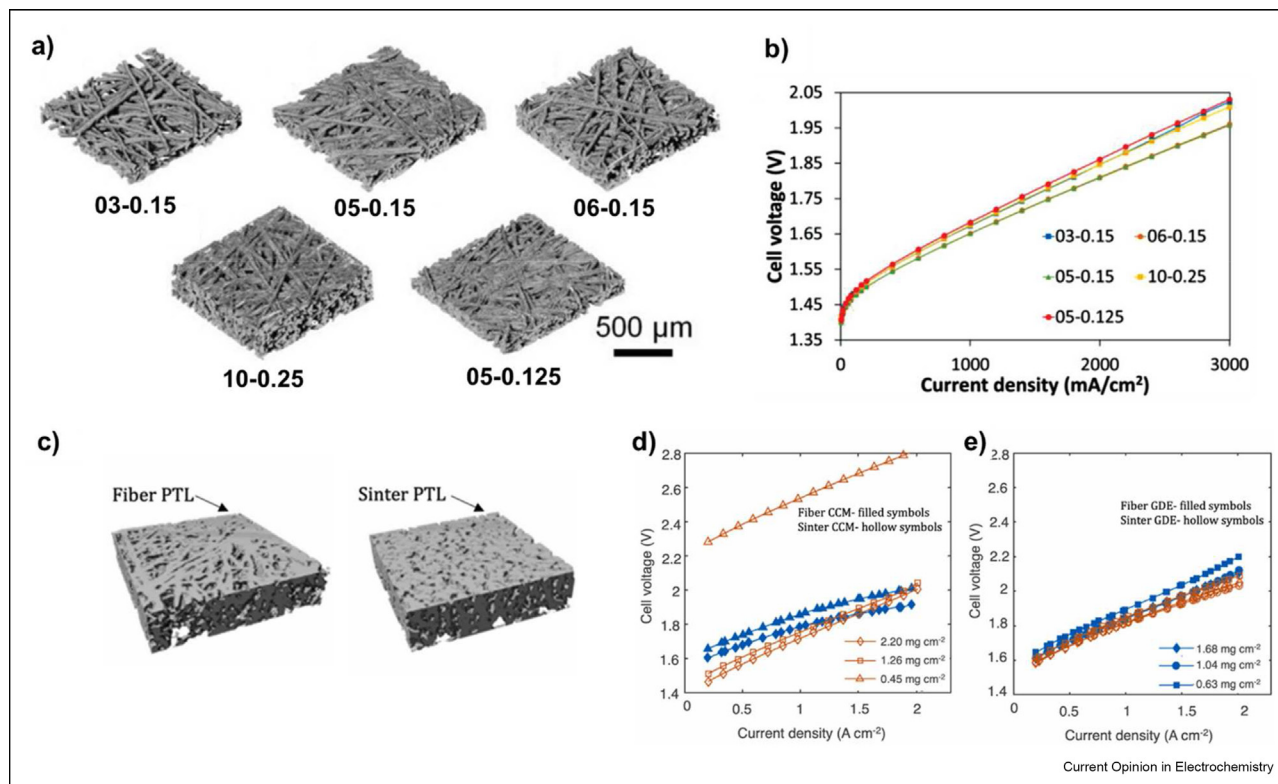
In addition, the PTL structural properties such as thickness, tortuosity, porosity, and pore size distribution will be critical to PEM-URFC performance [30]. Park et al. compare the PEM-URFC performance among three commercial carbon-based GDL with thicknesses ranging from 270 μm to 420 μm and demonstrate that GDL with moderate thickness attains the highest

RTE [24]. Optimization of the corrosion-resistant Ti-based PTLs using a similar approach can be promising to advance PEM-URFC performance as well. PTLs are available in various morphologies but are primarily studied for WE systems to date (Figure 3). Further research is desired to assess the bifunctionality of these PTLs in a PEM-URFC. In-depth analysis of the effects of structural properties of PTLs on PEM-URFC performance will be valuable to offer insights in materials design principles [31,32].

Membranes for PEM-URFCs

Membrane selection and design for a PEM-URFC is similar to that of a WE where thicker membranes (>120 μm) are necessary to ensure low gas crossover and sufficient mechanical stability [34–37] while thinner membranes (<30 μm) are preferred to minimize ohmic loss during both FC and WE operation [38,39]. Moreover, similar to a WE device that high differential pressure is applied to produce pressurized H_2 , such an operating condition is likely to be encountered in PEM-URFC charging mode, indicating the necessities to evaluate gas crossover and mechanical stability under differential pressure operations during PEM-URFC membrane design. In general, the ideal membrane for PEM-URFC should be thin and effectively confine gas crossover for safe operation and high efficiency. However, thinner membranes tend to have higher gas crossover. A potential solution is to modify thin membranes by incorporating a gas recombination layer (GRC), which typically contains a small amount of Pt or PtCo particles, into the membrane to allow the permeated H_2 to react with O_2 within the membrane. This strategy has been studied in the WE system in recent years as introduced by several reviews [40,41]. Integration methods include synthesizing a Pt-doped membrane [42], adding a Pt recombination interlayer between two membranes [43,44], and mixing the recombination PtCo catalyst with anode catalyst ink to obtain a recombination CCM [45,46]. Briguglio and coworkers show that their GRC design effectively minimizes H_2 crossover under differential pressure of up to 20 bar while using a relatively thin membrane of 90 μm [46]. However, to our knowledge that the potential application of GRC has not been explored in any PEM-URFC systems. In addition, membrane water management becomes more challenging in PEM-URFC as water must be drained when switching from WE to FC mode while avoiding membrane dehydration at the same time [30]. Fornaciari et al. propose a vapor-fed PEM-URFC to mitigate the water management issues during mode-switching [7]. Three membranes with the same thickness but different equivalent weights (EW) (NR212: EW1100, Aquivion E98-05S: EW980, Aquivion E87-05S: EW870) are investigated. NR212 shows the best performance in both WE and FC modes, which contradicts the expectation that lower EW should

Figure 3



PTLs with different morphologies and their performances are measured in WE devices. **a)** 3D rendering from X-ray computed tomography (XCT) images of five fiber PTLs labeled as 03–0.15, 05–0.15, 06–0.15, 10–0.25, and 05–0.125. Morphology properties are available in Ref. [31]. **b)** Polarization curves measured from PEM water electrolyzers using PTLs in a). **c)** 3D rendering of fiber and sinter PTLs. Detailed structural characterizations are available in Ref. [33]. Comparisons of polarization curves measured from PEM water electrolyzers using the fiber PTL and sinter PTL are studied with **d)** catalyst-coated membrane (CCM) and **e)** gas diffusion electrode (GDE) under various catalyst loadings. Adapted with permission from Ref. [31,33].

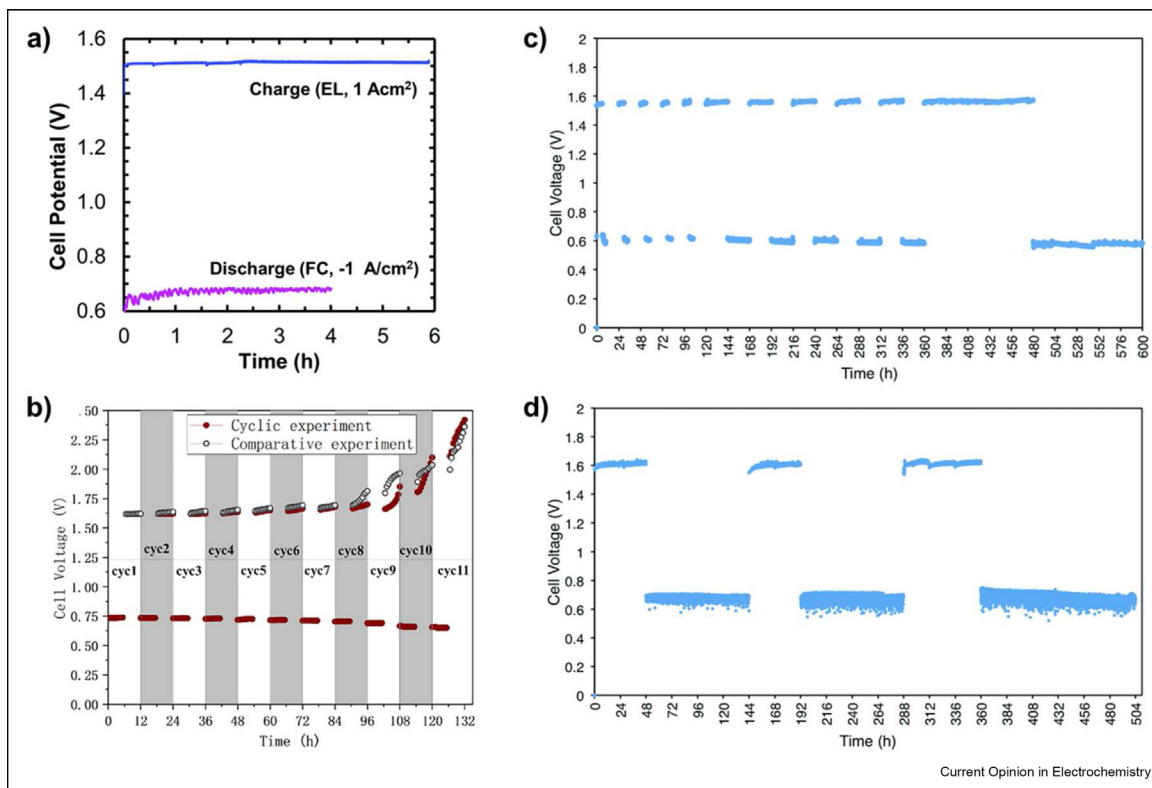
improve membrane hydration and higher protonic conductivity. The authors suggest that the discrepancies may arise from the membrane swelling effect and the mass transport loss at the ionomer/membrane interface for the Aquivion systems.

URFC durability evaluation

Typically, PEM-URFC durability is evaluated at the component or materials level, with a primary focus on catalyst degradation and carbon corrosion [20]. Common assessment criteria include comparing voltages or RTEs at targeted current densities (usually $<0.5 \text{ A/cm}^2$) after accelerated stress tests (ASTs) cycles [18,24] and mode-switching (reversible operation) cycles [11,21,25,47] or evaluating catalyst roughness [27] and dissolution rate [13] using cyclic voltammetry measurements. However, with growing interest in deploying PEM-URFC for large-scale applications such as grid balancing, it is essential to demonstrate reliable operation under charge-discharge operation at high current densities ($\geq 1.0 \text{ A/cm}^2$) for longer durations. Therefore, evaluating URFC durability at MEA level is more relevant. Li and coworkers

perform multiple 6-h-discharging and 6-h-charging cycles at 1 A/cm^2 with CE operation for 132 h using an MEA with carbon-based GDL (Figure 4a). The authors identify the carbon corrosion of GDL as the main cause of the increased ohmic and mass transport resistance [48]. Regmi *et al.* show that the stability of their MEA design can be maintained during 6 h of charging at 1 A/cm^2 followed by 4 h of discharging at 1 A/cm^2 , which can be analogous to a one-day operation scenario (Figure 4b) [21]. Peng *et al.* also conduct two longevity tests with up to 600 h of continuous reversible operation to evaluate durability of their MEA design (Figure 4c–d) [5]. Future research in this field needs to focus more on durability, and potentially even demonstrate H_2 production at elevated pressure during charging. These are critical steps to bring PEM-URFC technologies from laboratory research to markets for long-duration energy storage applications. On the other hand, research efforts are needed to design PEM-URFC ASTs protocols for fast materials screening (e.g., potential cycling for catalysts or electrodes) and component durability studies (e.g., pressure cycling for membranes). The correlation between ASTs and actual

Figure 4



Durability test of PEM-URFCs: **a)** demonstration of a PEM-URFC operating in a duck-curve duty cycle in CE configuration with a balanced 1 A/cm^2 charge and subsequent discharge (air), **b)** PEM-URFC cell voltage at 1 A/cm^2 during the cyclic experiment (red dotted line) and comparative experiment (black dotted line), **c)** durability test of CE mode PEM-URFC with charge/discharge at 1 A/cm^2 , **d)** durability test of CG mode PEM-URFC with charge and discharge conducted at 1 A/cm^2 and 0.5 A/cm^2 , respectively. Adapted with permission from Ref. [5,6,48]. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

URFC lifetime needs to be established for the fidelity of ASTs protocols.

Conclusions and prospects

To fully unleash the potential of PEM-URFCs as a clean, efficient, and reliable energy storage device for prolonged usage at the grid level, it is imperative to enhance cell performance at the electrode level while considering practical operating conditions. Prospective research efforts should involve optimizing each cell component along with developing effective operating strategies, such as using the CE configuration. New bifunctional catalyst materials will certainly be necessary to make future advances, but designing bifunctional electrode structure is perhaps more efficient to improve PEM-URFC performance and durability in the near term. The development of porous transport layers and membranes to balance both charging and discharging for high RTEs is required and can be crucial for the design of high-performing PEM-URFCs. We also recommend

that future studies evaluate cell performance at higher current densities (beyond kinetics region) and carry out durability tests under realistic operating conditions, such as long-term reversible operation.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Xiong Peng reports financial support was provided by US Department of Energy.

Data availability

Data will be made available on request.

Acknowledgements

The authors acknowledge the Department of Energy – Office of Energy Efficiency and Renewable Energy – Hydrogen and Fuel Cell Technologies Office (DOE-EERE-HFTO) for funding under Contract Number DE-AC02-05CH11231. Opinion presented in this work does not necessarily represent opinion by the U.S. Department of Energy.

References

Papers of particular interest, published within the period of review, have been highlighted as:

* of special interest

** of outstanding interest

- Chludzinski P, Danzig I, Fickett A, Craft D: *Regenerative fuel cell development for satellite secondary power*. GENERAL ELECTRIC CO LYNN MASS DIRECT ENERGY CONVERSION PROGRAMS; 1973.
- Wynveen R, Schubert F: *Regenerative fuel cell study*. Cleveland, Ohio (USA): Final report, Life Systems, Inc.; 1972.
- Devi Renuka K, Bhat SD, Unni SM: *Unitized regenerative fuel cells: future of renewable energy research*. *Renewable Energy Technologies*; 2022:375–402.
- Sadhasivam T, Dhanabalan K, Roh S-H, Kim T-H, Park K-W, Jung S, Kurkuri MD, Jung H-Y: **A comprehensive review on unitized regenerative fuel cells: crucial challenges and developments**. *Int J Hydrogen Energy* 2017, **42**:4415–4433.
- Peng X, Taie Z, Liu J, Zhang Y, Peng X, Regmi YN, Fornaciari JC, Capuano C, Binny D, Kariuki NN: **Hierarchical electrode design of highly efficient and stable unitized regenerative fuel cells (URFCs) for long-term energy storage**. *Energy Environ Sci* 2020, **13**:4872–4881.
Developed more porous and less tortuous Pt-Ir electrode that achieved RTEs higher than 50% at 1 A/cm² with both CG and CE operations and RTEs were well retained after longevity tests.
- Regmi YN, Peng X, Fornaciari JC, Wei M, Myers DJ, Weber AZ, Danilovic N: **A low temperature unitized regenerative fuel cell realizing 60% round trip efficiency and 10000 cycles of durability for energy storage applications**. *Energy Environ Sci* 2020, **13**:2096–2105.
Found that URFCs operating in CE configuration perform significantly better than in CG configuration and achieved stable 57% and 60% RTEs with air and O₂ as reductant gases at 1 A/cm² by optimizing Pt-Ir electrode catalyst composite.
- Fornaciari JC, Garg S, Peng X, Regmi YN, Weber AZ, Danilovic N: **Performance and durability of proton exchange membrane vapor-fed unitized regenerative fuel cells**. *J Electrochem Soc* 2022, **169**, 054514.
Performed a parametric study of vapor-URFC components and operation conditions including membrane EW, feed RH, and oxygen-electrode carrier gas and found that highest RTEs were achieved with higher EW membrane and 100% RH.
- Paul B, Andrews J: **PEM unitised reversible/regenerative hydrogen fuel cell systems: state of the art and technical challenges**. *Renew Sustain Energy Rev* 2017, **79**:585–599.
- Pu Z, Zhang G, Hassanpour A, Zheng D, Wang S, Liao S, Chen Z, Sun S: **Regenerative fuel cells: recent progress, challenges, perspectives and their applications for space energy system**. *Appl Energy* 2021, **283**, 116376.
- Omrani R, Shabani B: **Review of gas diffusion layer for proton exchange membrane-based technologies with a focus on unitized regenerative fuel cells**. *Int J Hydrogen Energy* 2019, **44**:3834–3860.
- Lim A, Jeong H-Y, Lim Y, Kim JY, Park HY, Jang JH, Sung Y-E, Kim JM, Park HS: **Amphiphilic Ti porous transport layer for highly effective PEM unitized regenerative fuel cells**. *Sci Adv* 2021, **7**, eabf7866.
Developed high-performance amphiphilic Ti PTL that improved both FC and WE performance simultaneously through a shadow mask patterning process using ultra-thin polydimethylsiloxane (PDMS) brush as a hydrophobic surface modifier.
- Rivera-Gavidia LM, de la Puente IF, Hernández-Rodríguez MA, Celorrio V, Sebastián D, Lázaro M, Pastor E, García G: **Bi-functional carbon-based catalysts for unitized regenerative fuel cells**. *J Catal* 2020, **387**:138–144.
- Kim J-H, Yun S-W, Shim K, You S-H, Jung S-M, Kweon H, Joo SH, Moon YH, Kim Y-T: **Enhanced activity and stability of nanoporous PtIr electrocatalysts for unitized regenerative fuel cell**. *ACS Appl Energy Mater* 2019, **3**:1423–1428.
- Khan M, Janjua NK, Ahmad A, Luque R, Al-Kahtani AA, Tighezza AM: **Highly effective bi-functional electrochemical activity of Ag₂O-PrO₂/γ-Al₂O₃ electrocatalysts towards OER and ORR**. *Int J Energy Res* 2022, **46**:14161–14173.
- Gochi-Ponce Y, Altamirano-Gutiérrez A, Fernández A: **Influence of the Pt/Ru electrocatalysts composition toward oxygen evolution and reduction reactions for unitized regenerative fuel cell**. *Int J Hydrogen Energy* 2021, **46**:26019–26026.
- Touni A, Papoulia C, Pavlidou E, Karfaridis D, Lambropoulou D, Sotiropoulos S: **(IrOx-Pt)/Ti bifunctional electrodes for oxygen evolution and reduction**. *Int J Hydrogen Energy* 2022, **47**:37811–37830.
- Klingenhof M, Hauke P, Brückner S, Dresch Sr, Wolf E, Nong HN, Spöri C, Merzdorf T, Bernsmeier D, Teschner D: **Modular design of highly active unitized reversible fuel cell electrocatalysts**. *ACS Energy Lett* 2020, **6**:177–183.
- Mo S, Jin C, Wang F, Wen C, Wang N, Feng L, Bai J, Liu J: **Nanophase-engineered bifunctional catalysts for oxygen electrodes in high-performance unitized regenerative fuel cells**. *ACS Appl Energy Mater* 2021, **4**:13361–13371.
- Mladenović D, Santos DM, Bozkurt G, Soylu GS, Yurtcan AB, Miljanić S, Šljukić B: **Tailoring metal-oxide-supported PtNi as bifunctional catalysts of superior activity and stability for unitized regenerative fuel cell applications**. *Electrochem Commun* 2021, **124**, 106963.
- Bhosale AC, Ghosh PC, Assaad L: **Preparation methods of membrane electrode assemblies for proton exchange membrane fuel cells and unitized regenerative fuel cells: a review**. *Renew Sustain Energy Rev* 2020, **133**, 110286.
- Lim A, Lee JS, Lee S, Lee SY, Kim H-j, Yoo SJ, Jang JH, Sung Y-E, Park HS: **Polymer electrolyte membrane unitized regenerative fuel cells: operational considerations for achieving high round trip efficiency at low catalyst loading**. *Appl Catal B Environ* 2021, **297**, 120458.
- Wang Y, Leung DY, Xuan J, Wang H: **A review on unitized regenerative fuel cell technologies, part-A: unitized regenerative proton exchange membrane fuel cells**. *Renew Sustain Energy Rev* 2016, **65**:961–977.
- Moriau LoJ, Bele M, Vižintin A, Ruiz-Zepeda F, Petek Ua, Jovanović P, Sala M, Gaberšček M, Hodnik N: **Synthesis and advanced electrochemical characterization of multifunctional electrocatalytic composite for unitized regenerative fuel cell**. *ACS Catal* 2019, **9**:11468–11483.
- Park JE, Karuppnanan M, Kwon OJ, Cho Y-H, Sung Y-E: **Development of high-performance membrane-electrode assembly in unitized regenerative fuel cells**. *J Ind Eng Chem* 2019, **80**:527–534.
- Lim A, Kim J, Lee HJ, Kim H-J, Yoo SJ, Jang JH, Park HY, Sung Y-E, Park HS: **Low-loading IrO₂ supported on Pt for catalysis of PEM water electrolysis and regenerative fuel cells**. *Appl Catal B Environ* 2020, **272**, 118955.
- Lee B-S, Park H-Y, Cho MK, Jung JW, Kim H-J, Henkensmeier D, Yoo SJ, Kim JY, Park S, Lee K-Y: **Development of porous Pt/IrO₂/carbon paper electrocatalysts with enhanced mass transport as oxygen electrodes in unitized regenerative fuel cells**. *Electrochem Commun* 2016, **64**:14–17.
- Kůš P, Ostroverkh A, Khalakhan I, Fiala R, Kosto Y, Šmíd B, Lobko Y, Yakovlev Y, Nováková J, Matolínová I: **Magnetron sputtered thin-film vertically segmented Pt-Ir catalyst supported on TiC for anode side of proton exchange membrane unitized regenerative fuel cells**. *Int J Hydrogen Energy* 2019, **44**:16087–16098.
- Omrani R, Shabani B: **Can PTFE coating of gas diffusion layer improve the performance of URFCs in fuel cell-mode?** *Energy Proc* 2019, **160**:574–581.
- Zhang Z, Guo M, Yu Z, Yao S, Wang J, Qiu D, Peng L: **A novel cooperative design with optimized flow field on bipolar plates and hybrid wettability gas diffusion layer for proton exchange membrane unitized regenerative fuel cell**. *Energy* 2022, **239**, 122375.

30. Vincent I, Lee E-C, Kim H-M: **Solutions to the water flooding problem for unitized regenerative fuel cells: status and perspectives.** *RSC Adv* 2020, **10**:16844–16860.
31. Peng X, Satjaritanun P, Taie Z, Wiles L, Keane A, Capuano C, Zenyuk IV, Danilovic N: **Insights into interfacial and bulk transport phenomena affecting proton exchange membrane water electrolyzer performance at ultra-low iridium loadings.** *Adv Sci* 2021, **8**, 2102950.
32. Leonard E, Shum AD, Danilovic N, Capuano C, Ayers KE, Pant LM, Weber AZ, Xiao X, Parkinson DY, Zenyuk IV: **Interfacial analysis of a PEM electrolyzer using X-ray computed tomography.** *Sustain Energy Fuels* 2020, **4**:921–931.
33. Kulkarni D, Huynh A, Satjaritanun P, O'Brien M, Shimpalee S, Parkinson D, Shevchenko P, DeCarlo F, Danilovic N, Ayers KE: **Elucidating effects of catalyst loadings and porous transport layer morphologies on operation of proton exchange membrane water electrolyzers.** *Appl Catal B Environ* 2022, **308**, 121213.
34. Carmo M, Fritz DL, Mergel J, Stolten D: **A comprehensive review on PEM water electrolysis.** *Int J Hydrogen Energy* 2013, **38**:4901–4934.
35. Arthurs C, Kusoglu A: **Compressive creep of polymer electrolyte membranes: a case study for electrolyzers.** *ACS Appl Energy Mater* 2021, **4**:3249–3254.
36. Danilovic N, Ayers KE, Capuano C, Renner JN, Wiles L, Pertoso M: **(Plenary) challenges in going from laboratory to megawatt scale PEM electrolysis.** *ECS Trans* 2016, **75**:395.
37. Ayers KE, Anderson EB, Capuano C, Carter B, Dalton L, Hanlon G, Manco J, Niedzwiecki M: **Research advances towards low cost, high efficiency PEM electrolysis.** *ECS Trans* 2010, **33**:3.
38. Peron J, Mani A, Zhao X, Edwards D, Adachi M, Soboleva T, Shi Z, Xie Z, Navessin T, Holdcroft S: **Properties of Nafion® NR-211 membranes for PEMFCs.** *J Membr Sci* 2010, **356**:44–51.
39. Breitwieser M, Klingele M, Vierrath S, Zengerle R, Thiele S: **Tailoring the membrane-electrode interface in PEM fuel cells: a review and perspective on novel engineering approaches.** *Adv Energy Mater* 2018, **8**, 1701257.
40. Ouimet RJ, Ebaugh TA, Mirshekari G, Bliznakov S, Bonville LJ, Maric R: **Current status on the manufacturing of nano-materials for proton exchange membrane energy systems by vapor-based processes.** *Energy Fuels* 2021, **35**:1933–1956.
41. Pham CV, Escalera-López D, Mayrhofer K, Cherevko S, Thiele S: **Essentials of high performance water electrolyzers—from catalyst layer materials to electrode engineering.** *Adv Energy Mater* 2021, **11**, 2101998.
42. Garbe S, Babic U, Nilsson E, Schmidt TJ, Gubler L: **Communication—pt-doped thin membranes for gas crossover suppression in polymer electrolyte water electrolysis.** *J Electrochem Soc* 2019, **166**:F873.
43. Mirshekari G, Ouimet R, Zeng Z, Yu H, Bliznakov S, Bonville L, Niedzwiecki A, Capuano C, Ayers K, Maric R: **High-performance and cost-effective membrane electrode assemblies for advanced proton exchange membrane water electrolyzers: long-term durability assessment.** *Int J Hydrogen Energy* 2021, **46**:1526–1539.
44. Klose C, Trinke P, Böhm T, Bensmann B, Vierrath S, Hanke-Rauschenbach R, Thiele S: **Membrane interlayer with Pt recombination particles for reduction of the anodic hydrogen content in PEM water electrolysis.** *J Electrochem Soc* 2018, **165**:F1271.
45. Pantò F, Siracusano S, Briguglio N, Aricò AS: **Durability of a recombination catalyst-based membrane-electrode assembly for electrolysis operation at high current density.** *Appl Energy* 2020, **279**, 115809.
46. Briguglio N, Siracusano S, Bonura G, Sebastián D, Aricò AS: **Flammability reduction in a pressurised water electrolyser based on a thin polymer electrolyte membrane through a Pt-alloy catalytic approach.** *Appl Catal B Environ* 2019, **246**:254–265.
47. Liu JX, Guo H, Ye F, Ma CF: **Cell voltage and two-phase flow in a unitized regenerative fuel cell operating in alternate fuel cell and electrolytic cell modes.** *Int J Green Energy* 2022:1–7.
48. Qiu D, Peng L, Shen S, Lai X: **Analysis of degradation mechanism in unitized regenerative fuel cell under the cyclic operation.** *Energy Convers Manag* 2022, **254**, 115210.
- Proposed a durability measurement protocol for URFC that could identify degradation mechanism at the cell level.