

Energetic Compromise in Small-Scale H₂ Energy Storage: A Comparative Experimental Study Based on Electrolyzers' Separators and Architectures [†]

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Abstract

The design of efficient small-scale hydrogen energy storage systems requires balancing hydrogen production rate, electrical efficiency, and system simplicity. This study experimentally investigates the energetic compromise imposed by separator material and electrolyzer architecture through a comparative analysis of finite-gap alkaline, finite-gap acidic, and zero-gap proton exchange membrane (PEM) electrolyzers. Zirfon[®] Pearl 500 (Agfa, Mortsel, Belgium) diaphragms were employed in alkaline electrolysis using 25 wt.% KOH, whereas Nafion[™] 117 (Chemours, Wilmington, DE, USA) membranes were used in both finite-gap acidic electrolysis (2.55 M H₂SO₄) and a commercial five-cell zero-gap PEM electrolyzer supplied with deionized water. Electrochemical performance was evaluated in terms of polarization behavior, apparent resistance, hydrogen production rate, Faradaic efficiency, and energy conversion efficiency. The zero-gap PEM architecture exhibited the best electrochemical performance, with an apparent resistance of only 0.138 Ω per cell, corresponding to reductions of approximately 43-, 51-, and 64-fold compared with the finite-gap PEM, stainless steel/Zirfon alkaline, and nickel/Zirfon alkaline configurations, respectively. The zero-gap electrolyzer delivered currents from 1.53 to 10.0 A while operating below 2.8 V, demonstrating the benefit of minimizing the ionic transport path. In contrast, the finite-gap acidic configuration achieved higher hydrogen production rates than the alkaline system owing to the superior proton conductivity of Nafion[™] 117, whereas the alkaline Ni/Zirfon configuration reached the highest Faradaic efficiency (≈98%) and energy conversion efficiency (≈36%) because of improved gas separation and reduced hydrogen crossover. Electrochemical impedance spectroscopy further revealed that the normalized ohmic resistance of the zero-gap PEM cell was only 0.029 Ω, with charge-transfer processes accounting for approximately 96.2% of the total impedance. These results demonstrate that separator properties and cell architecture govern the trade-off between reaction kinetics and energy efficiency, providing practical guidelines for selecting electrolyzer configurations dedicated to decentralized and small-scale hydrogen energy storage.



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Keywords: ionic transport path; apparent internal resistance; charge-transfer impedance; proton exchange membrane electrolyzer; alkaline electrolysis; zero-gap architecture; energy efficiency

1. Introduction

Hydrogen is increasingly recognized as a versatile energy carrier for supporting carbon-neutral energy systems because of its high gravimetric energy density and its ability to store electrical energy generated from intermittent renewable sources. Unlike fossil fuels, hydrogen can provide an energy-storage and energy-conversion pathway without direct carbon emissions at the point of use when produced from water using renewable electricity. This characteristic makes hydrogen particularly relevant for decoupling renewable electricity generation from energy demand and for addressing the temporal variability of solar and wind resources. Water electrolysis therefore represents an important technology for converting surplus renewable electricity into a storable chemical energy carrier and for enabling energy storage and sector coupling [1–3].

Among the available electrolysis technologies, alkaline water electrolysis (AWE) and proton exchange membrane water electrolysis (PEMWE) are the most mature. Alkaline electrolyzers typically employ concentrated potassium hydroxide solutions separated by porous diaphragms such as Zirfon[®], whereas PEM electrolyzers use proton-conducting membranes, commonly Nafion[®], which provide excellent ionic conductivity while preventing gas crossover [4–6].

The electrochemical performance of an electrolyzer depends not only on catalyst activity but also on separator properties, electrolyte conductivity, electrode material, and cell architecture. These parameters determine activation, ohmic, and mass transport losses, ultimately affecting hydrogen production rate, Faradaic efficiency, and overall energy conversion efficiency [7].

At laboratory scale, finite-gap (H-cell) configurations are extensively used because of their simplicity and flexibility for material screening. However, the relatively long ionic transport distance introduces significant ohmic losses that limit performance. In contrast, zero-gap architectures minimize the ionic path length by placing the electrodes directly against the membrane, substantially reducing transport resistance and improving current density [4,8]. Electrochemical impedance spectroscopy (EIS) provides an additional diagnostic approach for separating contributions associated with ohmic transport, interfacial charge transfer, and other dynamic polarization processes. In complex electrochemical energy systems, impedance-based diagnostics can also support condition monitoring and the identification of dynamic losses under multi-frequency excitation. Recent developments have demonstrated the potential of advanced online EIS approaches for monitoring multi-module electrochemical systems and managing interference associated with simultaneous impedance measurements [9]. In the present study, EIS was therefore used to provide complementary impedance information for the zero-gap PEM configuration, while the apparent resistance of the finite-gap cells was estimated from the linear region of their polarization curves. In addition to ionic conductivity and gas-separation capability, separator selection must consider the balance between ion-transport functionality and long-term chemical and structural stability. For polymeric ion-conducting membranes, parameters such as ion-exchange capacity, degree of functionalization, water uptake, mechanical integrity, and chemical stability can interact strongly with proton conductivity and durability. Increasing the density of ion-conducting functional groups may improve ionic transport, but excessive functionalization or hydration can also affect dimensional and chemical stability. Conse-

quently, the energetic performance of a separator should be considered together with its long-term stability rather than evaluated solely by its instantaneous conductivity or gas permeability [10].

Although numerous studies have independently investigated separator materials and electrolyzer architectures, relatively few have experimentally compared the energetic trade-off between finite-gap alkaline and PEM systems under comparable operating conditions. Understanding this compromise is essential for selecting appropriate electrolyzer configurations for decentralized and small-scale hydrogen production.

Therefore, this work experimentally compares finite-gap alkaline and acidic electrolyzers employing Zirfon® Pearl 500 diaphragms and Nafion™ 117 membranes, respectively, together with a commercial zero-gap PEM electrolyzer. The study evaluates polarization behavior, apparent resistance, hydrogen production rate, Faradaic efficiency, and energy conversion efficiency to identify the energetic compromise imposed by separator properties and electrolyzer architecture.

2. Materials and Methods

2.1. Electrolyzers' Configurations

Three electrolysis systems were experimentally investigated to evaluate the influence of separator material and cell architecture on hydrogen production performance. Two laboratory-scale finite-gap (H-cell) electrolyzers were assembled for alkaline and acidic electrolysis, while a commercial five-cell zero-gap PEM electrolyzer was used as a reference architecture as shown in Figure 1a,b.

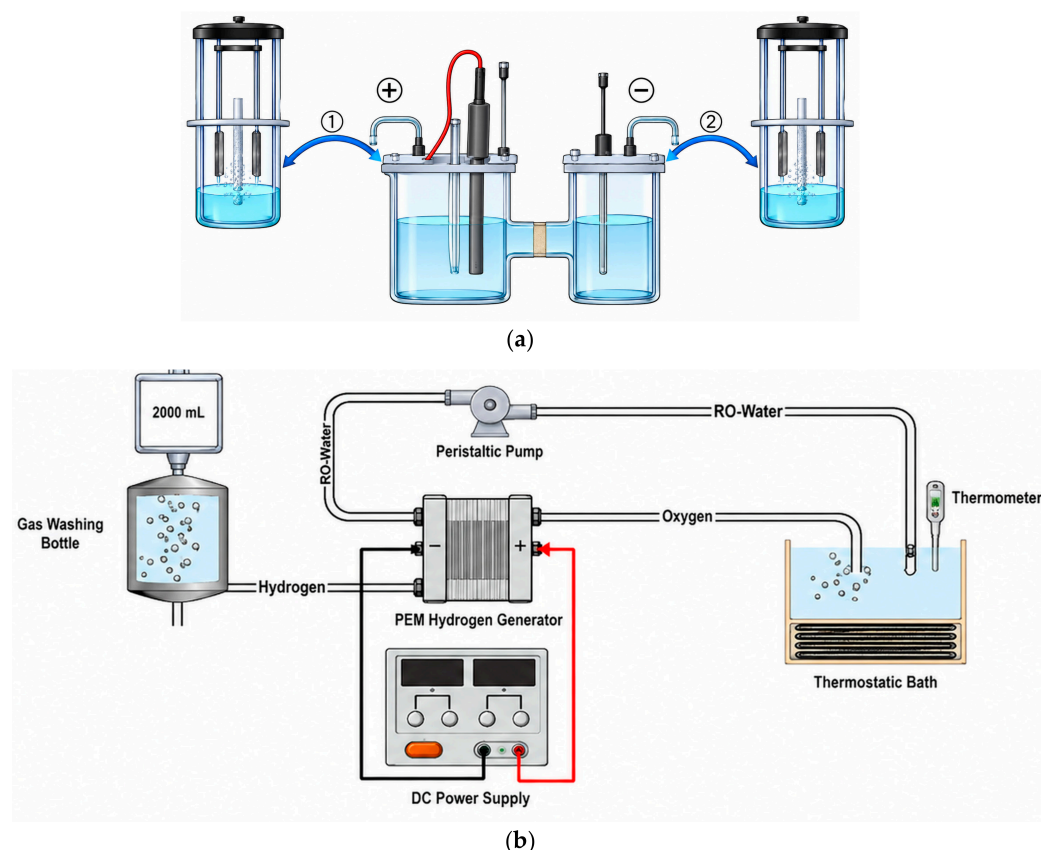


Figure 1. Experimental setups of the finite-gap (a) and zero-gap (b) configurations.

The alkaline H-cell employed a 25 wt% KOH electrolyte separated by a Zirfon® Pearl 500 diaphragm. Both nickel and stainless-steel plate electrodes with an active surface area

of 13.52 cm² were investigated. The electrode-to-diaphragm distance was fixed at 2 cm on each side.

The acidic H-cell used a 2.55 M H₂SO₄ electrolyte with a Nafion™ 117 membrane. Nickel electrodes identical to those employed in the alkaline system were used to allow meaningful comparison of separator effects.

The zero-gap PEM electrolyzer consisted of a five-cell stack operating with deionized water supplied directly to the membrane electrode assembly. Titanium felt and titanium mesh electrodes were integrated with Nafion™ 117 membranes to minimize ionic transport distance.

The detailed configurations of the previous systems are presented in Table 1.

Table 1. Detailed configurations of the electrolyzers.

Parameter	Alkaline Electrolysis	Acidic Electrolysis	Zero-Gap PEM Electrolysis
Electrolyte type	KOH	H ₂ SO ₄	Solid Polymer Electrolyte (SPE), deionized water feed
Electrolyte concentration	25 wt.% (4.46 M)	22 wt.% (2.55 M)	— (solid electrolyte)
Electrodes' material and structure	Nickel/Stainless-Steel-304 plates	Nickel plates	Titanium mesh and titanium felt
Anode cross-sectional area	13.5 × 2 cm ² (27 cm ²)	13.5 × 2 cm ² (27 cm ²)	20 cm ²
Cathode cross-sectional area	13.5 × 2 cm ² (27 cm ²)	13.5 × 2 cm ² (27 cm ²)	20 cm ²
Distance from anode to membrane	2 cm	2 cm	0 cm (zero-gap configuration)
Distance from cathode to membrane	2 cm	2 cm	0 cm (zero-gap configuration)
Electrolyte/membrane ionic conductivity	602.3 mS/cm	600 mS/cm	Nafion™ 117 proton conductivity: 80–100 mS/cm (ambient temperature)
Applied cell potential	4 V	4 V	1.5–2.8 V
Type of separator/membrane	Zirfon® Pearl 500	Nafion™ 117	DuPont™ Nafion™ 117 proton exchange membrane
Electrolyzer architecture	Single-cell	Single-cell	5-cell zero-gap PEM stack
Active area per cell	27 cm ²	27 cm ²	20 cm ²
Bipolar plates material	—	—	Stainless Steel 304

2.2. Experimental Procedure

Polarization curves were obtained by progressively increasing the applied cell voltage while measuring the corresponding current response. Hydrogen production was determined experimentally using volumetric gas measurements and converted into mass flow rate. The experimentally collected gas volume was used to determine the hydrogen production rate and was compared with the theoretical hydrogen production calculated from Faraday's law. The present dataset was intended for comparative performance evaluation and did not include dedicated gas-chromatographic analysis of dissolved hydrogen or a separate quantitative determination of hydrogen crossover. Therefore, differences in Faradaic efficiency are interpreted as overall gas-production efficiency differences rather than as direct quantitative measurements of hydrogen crossover. Faradaic efficiency was calculated by comparing the experimentally measured hydrogen production with the theoretical production predicted by Faraday's law.

Energy conversion efficiency was evaluated from the ratio between the higher heating value of produced hydrogen and the supplied electrical energy. The apparent internal resistance of the finite-gap electrolyzers was estimated from the slope of the linear region of the polarization curves according to Ohm's law. These values are referred to as apparent resistances because the polarization-derived slope may include contributions from ohmic transport, electrode kinetics, and other polarization losses and should not be interpreted as a direct measurement of pure ohmic resistance. For the zero-gap PEM electrolyzer, electrochemical impedance spectroscopy was additionally performed to distinguish the ohmic contribution from the charge-transfer contribution.

For the zero-gap PEM stack, cyclic voltammetry and electrochemical impedance spectroscopy (Nyquist analysis) were additionally performed to separate ohmic resistance from charge-transfer resistance.

The performance of the investigated electrolyzers was evaluated using several key electrochemical and energy-related parameters, including polarization behavior, current density, apparent resistance, hydrogen production rate, Faradaic efficiency, and energy conversion efficiency. Together, these indicators provide a comprehensive assessment of both the electrochemical kinetics and the overall energy performance of the different electrolysis systems.

3. Results and Discussion

3.1. Influence of Electrode Material

Nickel electrodes consistently exhibited superior electrochemical performance compared with stainless-steel electrodes (Figure 2). The higher catalytic activity of nickel promoted faster hydrogen evolution kinetics, resulting in higher current densities under identical operating conditions. Although both electrode materials were evaluated in the same alkaline electrolyte and with identical cell geometry, the nickel electrode reduced activation losses without significantly affecting ionic transport through the electrolyte. Consequently, the Ni/Zirfon configuration reached higher hydrogen production rates while maintaining comparable transport characteristics.

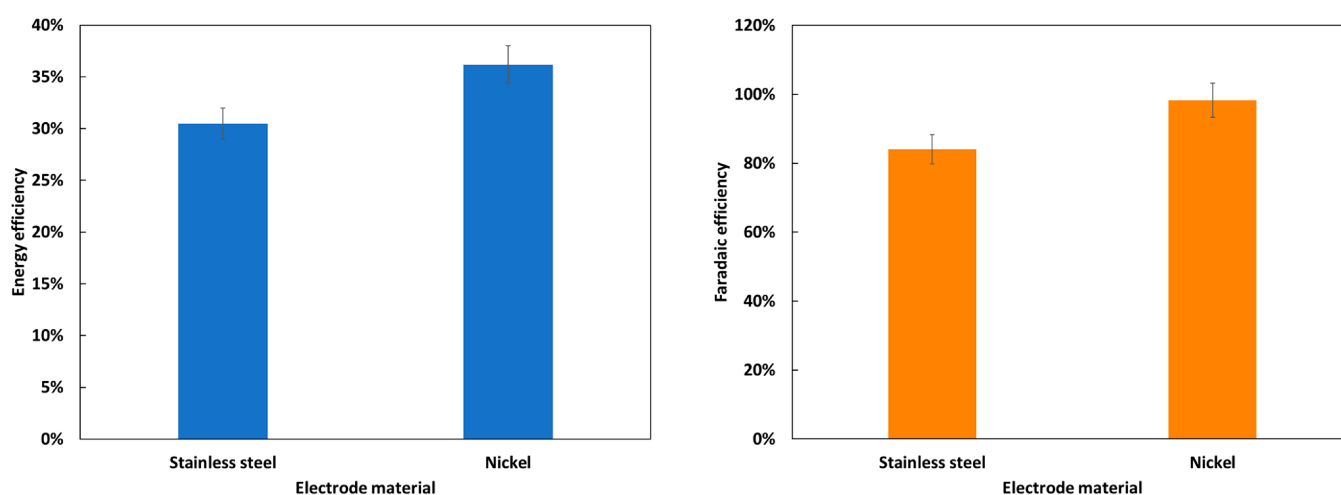


Figure 2. Influence of electrode material in terms of energy efficiency (left) and Faradaic efficiency (right).

The energetic analysis further indicates that the effect of electrode material is mainly kinetic rather than ohmic. Stainless-steel electrodes exhibited slower reaction kinetics, lower conductivity, and higher apparent resistance, whereas nickel electrodes reduced activation overpotential and improved charge-transfer processes, thereby directing a larger fraction

of the supplied electrical energy toward hydrogen evolution (Figure 2). These observations agree with previous studies identifying nickel as one of the most active and economically attractive non-noble HER catalysts for alkaline electrolysis owing to its favorable adsorption energy and excellent electrochemical stability [6,7,9–11].

3.2. Effect of Separator Material

Separator properties strongly affected both electrochemical kinetics and energetic performance (Figure 3). The acidic H-cell equipped with a Nafion™ 117 membrane exhibited an apparent resistance of approximately 6.0 Ω , whereas the alkaline configuration employing a Zirfon® Pearl 500 diaphragm presented resistances between 7.05 Ω (stainless-steel electrodes) and 8.83 Ω (nickel electrodes). The higher proton conductivity of Nafion facilitated faster ionic transport and consequently produced substantially higher current densities and hydrogen production rates. These observations are consistent with numerous studies reporting the high proton conductivity and low area-specific resistance of Nafion membranes in PEM electrolyzers [4,5,12,13]. The acidic H-cell employed nickel electrodes in 2.55 M H₂SO₄. The results obtained with this configuration are therefore interpreted as the electrochemical response of the selected electrode/electrolyte combination under the investigated conditions. The discussion does not extend these short-term observations to broader conclusions about nickel durability in strongly acidic media.

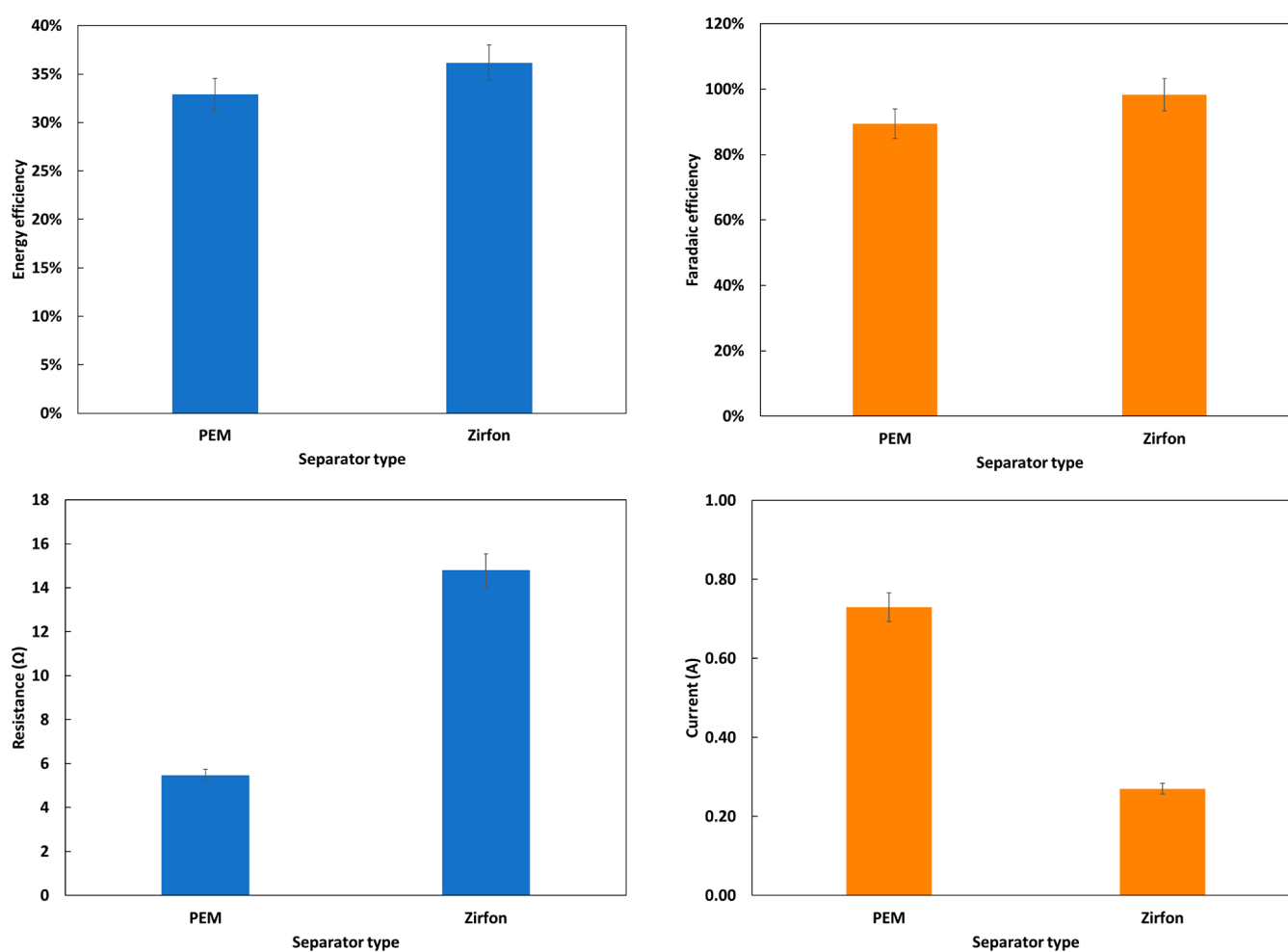


Figure 3. Effect of separator material in terms of energy efficiency (upper left), Faradaic efficiency (upper right), resistance (lower left), and current (lower right).

Conversely, the Zirfon diaphragm is designed to provide effective gas separation, which can contribute to reduced gas crossover and parasitic recombination. In the present study, however, the higher Faradaic efficiency observed for the alkaline Zirfon configuration is therefore discussed as an overall gas-production performance difference between the investigated configurations. As shown in Figure 3, the alkaline Zirfon configuration achieved the highest measured Faradaic efficiency (approximately 98%) together with an energy conversion efficiency of approximately 36%, despite its lower current output than the acidic Nafion configuration. This result indicates that hydrogen production efficiency and electrical conversion efficiency are not necessarily maximized by the same separator configuration. These findings agree with previous investigations showing that Zirfon diaphragms exhibit excellent chemical stability, low gas permeability, and efficient gas separation, thereby improving Faradaic efficiency during alkaline water electrolysis [6,7,14].

The experimental comparison therefore reveals a clear energetic compromise. Although the Nafion membrane minimizes ohmic losses through its superior proton conductivity, the Zirfon diaphragm converts electrical energy into hydrogen more efficiently because of its lower hydrogen crossover and improved gas-separation capability. These results demonstrate that separator selection simultaneously governs ionic conductivity, gas purity, Faradaic efficiency, hydrogen production rate, and overall energy efficiency [4–7,12–14].

3.3. Effect of Electrolyzer's Architecture

The zero-gap PEM architecture exhibited substantially lower apparent resistance than the finite-gap configurations (Figure 4). This comparison is interpreted primarily as an evaluation of the combined effect of electrolyzer architecture and ionic transport distance rather than as a geometry-independent comparison of intrinsic electrode or separator kinetics. In the finite-gap cells, the electrodes were positioned 2 cm from the separator on each side, whereas the zero-gap PEM configuration placed the electrodes directly against the membrane, minimizing the ionic transport path.

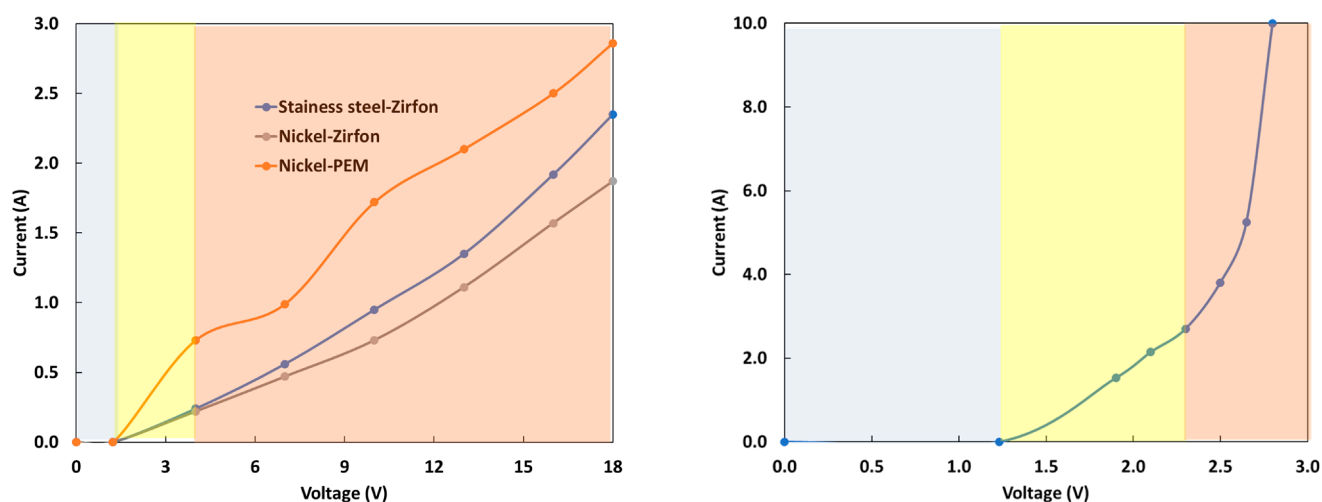


Figure 4. Effect of electrolyzer architecture in terms of polarization curves for finite-gap (left) and zero-gap (right) electrolyzers, blue, yellow and red zones refer to reversible, activation and ohmic potentials.

Linear fitting of the polarization curves yielded an apparent resistance of approximately 0.138Ω per cell for the zero-gap PEM electrolyzer, compared with approximately 6.00Ω for the finite-gap PEM configuration, 7.05Ω for the stainless-steel/Zirfon alkaline H-cell, and 8.83Ω for the nickel/Zirfon alkaline H-cell. Consequently, the zero-gap architecture exhibited apparent resistances approximately 43, 51, and 64 times lower than those

of the finite-gap PEM, stainless-steel/Zirfon, and nickel/Zirfon configurations, respectively. These differences demonstrate the strong influence of ionic transport distance and cell architecture on the measured electrochemical response [4,5,8,13,15].

The reduction in ohmic losses translated directly into lower operating voltages. A current of 1 A was obtained at only ≈ 1.67 V in the zero-gap PEM electrolyzer, whereas the finite-gap PEM, stainless-steel/Zirfon, and nickel/Zirfon configurations required approximately 7.04 V, 10.38 V, and 12.13 V, respectively. At 2 A, the zero-gap system operated at only ≈ 2.05 V, while the finite-gap PEM required ≈ 12.21 V, the stainless-steel/Zirfon cell ≈ 16.37 V, and the nickel/Zirfon cell did not reach this current within the investigated voltage range. These results clearly demonstrate that minimizing the ionic transport distance substantially reduces electrical energy consumption for a given hydrogen production rate, consistent with recent studies on compact PEM electrolyzers [4,5,13,15,16].

Furthermore, the zero-gap PEM stack delivered currents ranging from 1.53 to 10.0 A, corresponding to current densities of $0.076\text{--}0.500\text{ A cm}^{-2}$, with a fitted resistance of only $0.138\ \Omega$ per cell and a maximum power output of approximately 28 W per cell (≈ 140 W for the five-cell stack). These results demonstrate that cell architecture has a greater influence on electrochemical performance than electrolyte conductivity alone and confirm that minimizing transport losses is essential for achieving highly efficient hydrogen production systems [5,8,13,15,16].

These results demonstrate that minimizing the ionic transport distance substantially reduces the voltage required to reach a given current under the investigated operating conditions. However, because the tested systems differ in architecture, active area, electrode configuration, and operating conditions, the results should not be interpreted as a strict separation of intrinsic kinetic effects from architectural effects.

3.4. Electrochemical Characterization of Zero-Gap Electrolyzer

Electrochemical impedance spectroscopy provided complementary evidence regarding the internal electrochemical losses of the zero-gap PEM architecture (Figure 5). Because EIS measurements were performed only for the zero-gap PEM electrolyzer, the resulting ohmic and charge-transfer contributions cannot be directly compared with EIS-derived parameters for the finite-gap alkaline and acidic cells. Instead, the EIS results are used to characterize the internal impedance distribution of the zero-gap configuration. (Figure 5). After normalization to a single cell, the ohmic resistance was only $0.029\ \Omega$, whereas the total impedance reached $0.767\ \Omega$, indicating a charge-transfer resistance of approximately $0.738\ \Omega$. Consequently, only 3.8% of the total impedance originated from ohmic losses, while 96.2% resulted from charge-transfer processes occurring at the electrode–electrolyte interface. The predominance of charge-transfer impedance in the measured zero-gap configuration indicates that, once the ionic transport path is minimized, interfacial electrochemical processes become an important contribution to the measured impedance. This observation is consistent with the general application of EIS as a diagnostic technique for separating electrochemical loss mechanisms in electrochemical energy systems [4,5,8,9].

These results demonstrate that the zero-gap architecture effectively suppresses ionic transport losses by minimizing the electrode-to-membrane distance. Once ohmic losses become negligible, the overall energetic limitation shifts toward electrochemical reaction kinetics. Therefore, further improvements in zero-gap PEM electrolyzers should primarily focus on catalyst development, optimization of the membrane–electrode assembly, and enhancement of charge-transfer kinetics rather than additional reductions in ionic resistance. This conclusion is consistent with recent advances in PEM electrolyzer design and catalyst engineering [5,8,15–19].

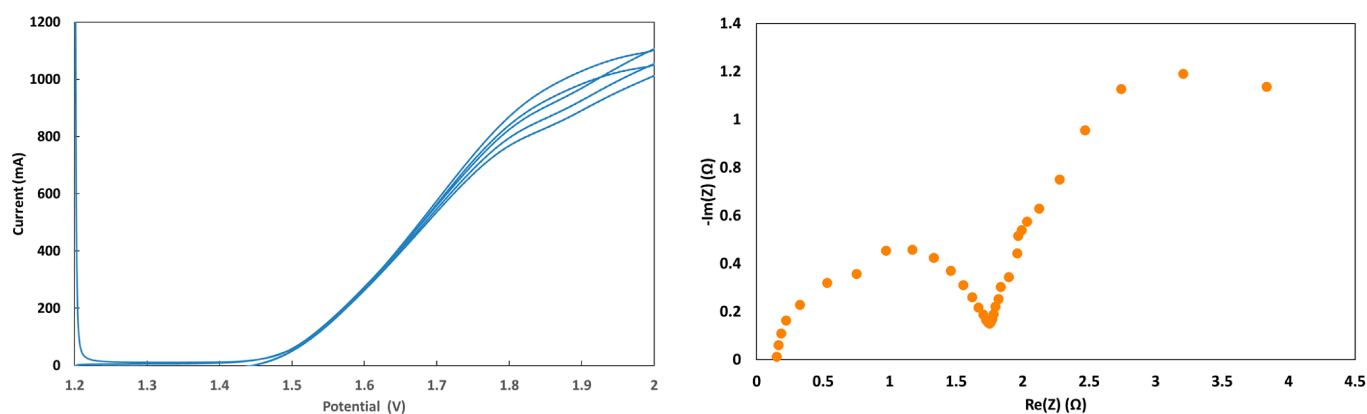


Figure 5. Cyclic voltammetry (left) and Nyquist impedance spectroscopy (right) of the zero-gap electrolyzer.

3.5. Limitations of the Experimental Comparison

Several limitations should be considered when interpreting the present results. First, the investigated electrolyzers differ in architecture, electrode-to-separator distance, active area, and electrolyte configuration. Consequently, the comparison quantifies the practical energetic consequences of the investigated configurations but does not constitute a geometry-independent determination of intrinsic catalyst or separator kinetics.

Second, the apparent resistance of the finite-gap electrolyzers was obtained from the linear region of the polarization curves, whereas EIS was performed only for the zero-gap PEM electrolyzer. Therefore, the polarization-derived values should be considered apparent resistances rather than direct measurements of pure ohmic resistance, and a complete equivalent-circuit comparison among all configurations would require additional EIS measurements.

Third, the acidic configuration employed nickel electrodes in 2.55 M H_2SO_4 . The present study did not include ICP analysis, post-test elemental analysis, or long-term chronopotentiometric measurements. Therefore, the results demonstrate the short-term electrochemical response of the investigated configuration but do not establish the long-term corrosion resistance or durability of nickel under these acidic conditions.

Fourth, hydrogen production was determined by volumetric gas measurements. Dedicated gas-chromatographic analysis of dissolved hydrogen and direct quantitative measurements of hydrogen crossover were not performed. Accordingly, differences in Faradaic efficiency are discussed as overall experimental gas-production efficiency differences rather than as direct quantitative measurements of crossover.

Finally, the present work focuses on short-term polarization and efficiency behavior and does not include multi-hour aging or degradation experiments. Long-term separator stability, electrode corrosion, membrane degradation, and performance retention should therefore be investigated in future work before definitive conclusions regarding long-term engineering lifetime are drawn.

4. Conclusions

This study experimentally compared finite-gap alkaline, finite-gap acidic, and zero-gap PEM electrolyzers to investigate the influence of separator properties and electrolyzer architecture on short-term electrochemical and energetic performance.

The results showed that electrode material mainly influenced the electrochemical response through kinetic effects. Nickel electrodes exhibited higher performance than stainless-steel electrodes in the investigated alkaline configuration, resulting in higher current densities and improved hydrogen production performance.

Separator selection produced a clear trade-off between hydrogen production rate and energy utilization. The Nafion™ 117 acidic configuration exhibited lower apparent resistance and higher current output than the Zirfon® Pearl 500 alkaline configuration, whereas the alkaline Zirfon configuration achieved the highest measured Faradaic efficiency (approximately 98%) and energy conversion efficiency (approximately 36%) among the investigated finite-gap systems.

The zero-gap PEM configuration exhibited the lowest apparent resistance, approximately 0.138 Ω per cell, and operated at substantially lower cell voltages than the finite-gap configurations. These results demonstrate the strong influence of ionic transport distance and cell architecture on electrochemical response. However, because the investigated systems differed in geometry, active area, electrode configuration, and operating conditions, the observed performance differences should not be interpreted as a geometry-independent comparison of intrinsic catalyst or separator kinetics.

EIS characterization of the zero-gap PEM configuration further showed a normalized ohmic resistance of approximately 0.029 Ω per cell, while charge-transfer processes represented the dominant contribution to the measured impedance. This result indicates that, after minimizing ionic transport losses through a zero-gap architecture, further performance improvements should increasingly focus on electrode kinetics, membrane-electrode interfaces, and charge-transfer processes.

The results presented in this study describe the short-term electrochemical and energetic behavior of the investigated configurations. Their interpretation is therefore centered on the measured performance trade-offs, while longer-term durability and degradation remain important considerations for future development. Therefore, the present results should be considered evidence of the short-term energetic and electrochemical trade-offs among the investigated configurations rather than proof of long-term electrode or separator durability.

Overall, the results demonstrate that optimizing small-scale hydrogen electrolyzers requires simultaneous consideration of ionic transport distance, separator properties, electrode kinetics, Faradaic efficiency, and electrical energy consumption. Future studies combining these performance indicators with durability, gas crossover, and impedance-monitoring assessments would provide a broader basis for evaluating the long-term performance of different configurations.

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