



Review Article

Advancements in hydrogen production using alkaline electrolysis systems: A short review on experimental and simulation studies

Lucía Paula Campo Schneider^{1,a}, Maryem Dhrioua^{1,a},
Dirk Ullmer¹, Franz Egert^{1,2}, Hans Julian Wiggenhauser¹,
Kamal Ghotia¹, Nicolas Kawerau¹, Davide Grilli¹,
Fatemeh Razmjooei¹ and Syed Asif Ansar¹



Abstract

Although alkaline water electrolysis (AWE) is a highly mature technology for hydrogen production, its potential is hindered by relatively low efficiencies at high current densities. On the other hand, to conform with “RePowerEU” directives, coupling electrolyzers with new renewable energy sources (RES) is highly demanded. However, integrating fluctuating RES poses challenges for the AWE due to increasing gas impurity as the current density decreases. Herein, we revised the most promising recent developments in materials, cell design, and system integration aimed at conquering the aforementioned challenges. It is shown that the implementation of advanced components and control strategies, e.g. electrolyte management, is vital to enhance the efficiency at high current densities and expand the load range of operation by maintaining the high gas purity.

Addresses

¹ Institute of Engineering Thermodynamics, German Aerospace Center, Pfaffenwaldring 38-40, Stuttgart 70569, Germany

² University of Stuttgart, Faculty 6 – Aerospace Engineering and Geodesy, Stuttgart 70569, Germany

Corresponding author: Corresponding author: Razmjooei, Fatemeh (fatemeh.razmjooei@dlr.de)

^a These authors have equal authorship contribution.

the key to reduce global CO₂ emissions from around 32 Gt in 2023 to less than 2 Gt in 2050 [1]. The need to reduce greenhouse gas (GHG) emissions has promoted the use of what is called “green H₂,” i.e. hydrogen produced by water electrolysis employing renewable power sources [2]. Green H₂ production is expected at first to substitute conventional H₂ produced from fossil sources in the chemical industry, but might then also become a major energy carrier potentially used for long- and short-term energy storage (chemical storage) to balance energy demand and production [3,4]. The high purity H₂ obtained by water electrolysis (>99.9%) can later be used for power generation through thermo-mechanical or electro-chemical convertors, synthetic fuels and other hydrogen derivatives production, and in the chemical industry [5,6]. According to the Net Zero Emissions by 2050 Scenario, rising hydrogen use is expected to reach from 95 Mt in 2022 to more than 660 Mt in 2050, increasing from 0.1% to 60% of the share of electrolysis (3000 GW electrolyzers) [7,8].

Alkaline water electrolysis (AWE) is a mature technology with a high technology readiness level (TRL 9 [9]) and represents 58% of the electrolyzer manufacturing capacity in Europe [10]. Thanks to its cost benefits, AWE presents a promising solution for large-scale hydrogen production [11]. By 2023, there was 81 MW_{el} of installed capacity of AWE and 1200 MW_{el} under construction [10]. Typically, its operation temperature ranges from 40°C to 80°C, and pressures reach up to 30 bar [4,12]. The entire system is designed for lifetimes greater than 60,000 h and high H₂ conversion efficiency, which currently is close to 70% considering the lower heat value of H₂ [3]. The coupling of renewable power energy sources (RES) with H₂ production is theoretically possible thanks to the electrolyzers electrical fast response [5], but historically the AWE systems, including balance of plant (BoP) equipment, have been designed for operation at steady-state conditions [4]. The limitation of current densities up to 0.2–0.6 A·cm⁻² [13,14] combined with a minimum

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Introduction

A complete transformation in the energy sector, i.e. energy production, transport, and consumption, holds

partial load of 10%–40% of the nominal load [15], restrict the ability of the AWE system to operate under the highly variable loads associated to RES. Furthermore, the slow thermal dynamic response at system level, which leads operating at lower temperatures to avoid component damage during fast load increase, reduces AWE efficiency. The main challenges identified for the AWEs, particularly when integrated with RES, are the low efficiency, low gas purity at low loads, high cell voltage at high current densities, long-term durability, cost, and the system's dynamic response [3,4,15]. Innovations could demonstrate AWE's potential to increase current density and hydrogen production while simultaneously decreasing costs [11].

In this context, researchers have directed their efforts toward enhancing and optimizing cell/stack material and configuration, adjustment of operation under dynamic, and partial loads for better upstream and downstream AWE integration. This study highlights the latest developments in AWEs technology and its integration into the energy system, with the specific focus on challenges associated with component level and operation under fluctuating load profiles and the prerequisites for downstream integration.

Developments at cell/stack level design and its components

In the following section, cell/stack and material designs for AWE are discussed, along with the most advanced practices. The commonly used stack configuration, series cells electrically connected by bipolar plates, is introduced in Figure 1(a–c) [14]. The cell is composed of two half-cells separated by a diaphragm or membrane to minimize the gas mixture, and each half cell includes an electrode, a gas diffusion layer to facilitate the bubble removal, and a bipolar plate. At the cell level, modifications of gap between electrodes, electrolyte, and gas flow inside the cell, electrodes and their microstructures, and separator properties have resulted in an increased durability and efficiency due to a decrease in the overall cell resistance, including ohmic and activation losses. As shown in Figure 1(d,e), at lower current densities, the cell performance is governed mainly by the catalytic activity, whereas at higher current densities, the ohmic losses related to separators and electrode microstructure play a major role [16]. In particular, electrodes, their porous transport layer's microstructure, and separator's conductivity impact the ohmic losses associated with mass transport, by affecting the ion transport to the reacting sites, and the bubble removal capacity [11].

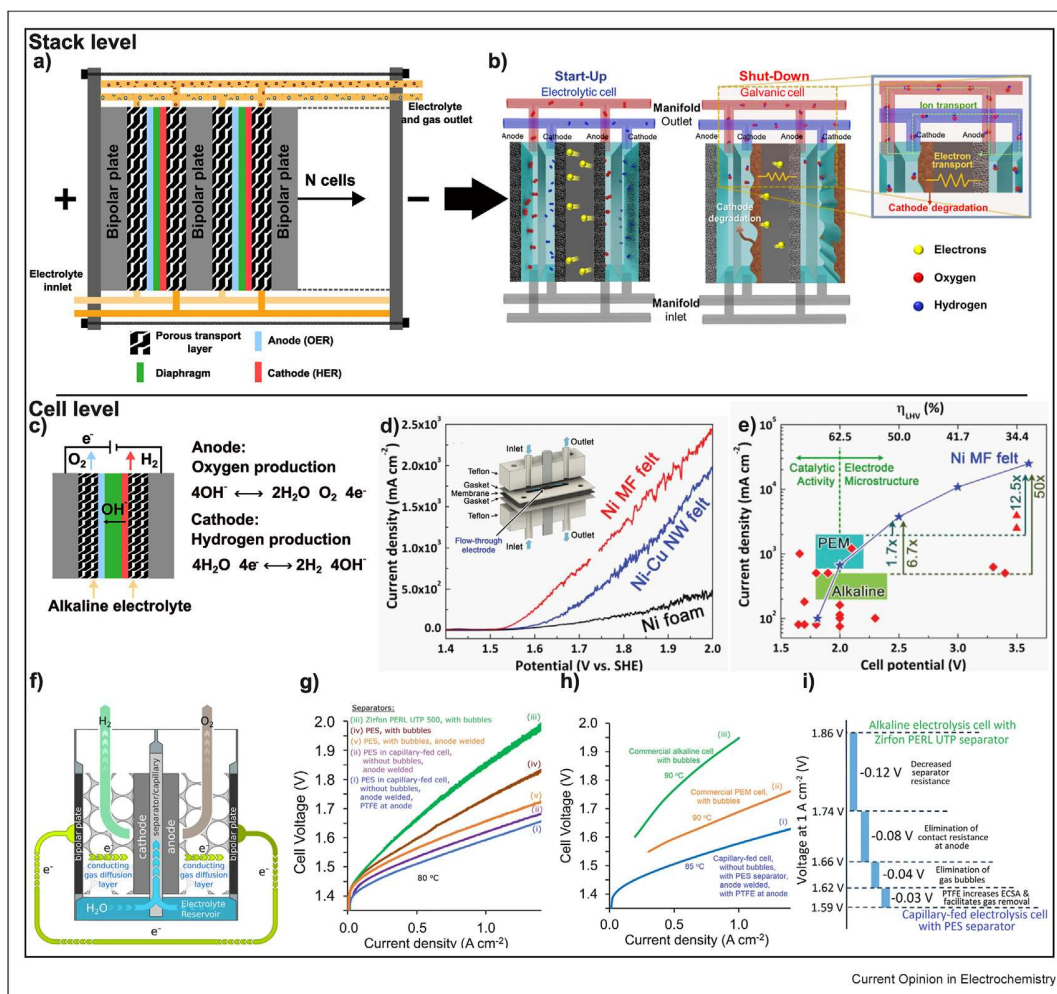
Precious metal-based catalysts demonstrated the highest activity for the hydrogen and oxygen evolution reaction (HER, OER). However, due to its scarcity and cost, they have been replaced by Ni-based electrodes. Table 1 summarizes recently developed catalysts for OER and

HER. It can be seen that comparison between different catalysts is not straightforward since there is not yet a harmonized protocol among scientific community for the performance characterization. Reviews on promising nonprecious metal catalyst developments such as Fe-(Ni or Co) for OER, transition metal nitrides, and Ni–Mo for HER can be found in Refs. [17,18] and [19].

Unsteady operation of the commonly used Ni-based catalysts can contribute to their deactivation due to microstructural and chemical changes [20]. Anode based on NiFe-hydroxide/oxide-coated stainless steel has shown a stable overpotential after 20,000 potential cycles from 0.5 to 1.8 V (vs. reference hydrogen electrode (RHE)), tested at 20 °C in 7 M KOH), while under the same conditions, pure Ni showed a 100 mV increase in overpotentials at 0.1 A·cm⁻² [20]. Self-repairing catalyst layers have been developed based on cobalt hydroxide nanosheets (Co-ns) with a tripodal ligand on Ni and were able to maintain their low overpotential with an increase of just 9 mV after 40,000 potential cycles (from 0.5 to 1.8 V vs. RHE). The repairing process occurred thanks to the addition of a dispersion of Co-ns to the 1 M KOH electrolyte, which were deposited on the Ni anode during the constant current phases of the test [21]. The durability of the electrodes is also affected by the transients during start-up and shut-down, which generate a reverse current flow during the off periods through the ionic transport provided by the electrolyte in the stack manifold (Figure 1(b)) and promote an irreversible oxidation of the Ni cathode to β-Ni(OH)₂ or NiO [22]. Efforts to limit this degradation phenomenon include the implementation of cathodic protection systems by sacrificial anodes [22] and the design of electrodes with sacrificial species incorporated into them [23].

The energy losses that arise from the formation of bubbles within electrolysis systems, by blocking the catalyst surface and increasing activation and ohmic loss [24], limit the maximum operating current density [25]. In a recent review on the effect of bubbles on system efficiency, it has been shown that at least 5%–10% of the electrolysis system energy can be saved by mitigating bubble formation and that strategies such as centrifuging the electrolyzer to apply a supergravity environment for a gas bubble-free operation can lead to an increase of 9%–17% of the stack energy efficiency (at current densities greater than 500 mA·cm⁻² and relative to the higher heating value of hydrogen) [25]. Novel systems, characterized by micro-structured porous electrodes [16] and capillary-fed alkaline electrolysis cells (Figure 1(f–i)) [26], hold the promise of a significant leap in energy efficiency thanks to a decrease in bubble accumulation or directly avoiding bubble formation. Studies on the effect of the microstructure of Ni electrodes operated in zero-gap flow-through single cell configuration (Figure 1(d,e)) showed the existence of an

Figure 1



Electrolyzer at stack and cell level. (a) Schematic representation of the stack components. (b) Degradation mechanisms during shut-down; along with the electrodes, the electrochemical circuit is completed by the bipolar plate (e-transport) and the electrolyte present in the stack manifold (ion transport). During operation, each cell acts as an electrolytic cell, but during shut down, a reverse current flow is generated, and the anode and cathode of two adjacent cells reverse their reactions and form a galvanic cell. This leads to the Ni electrode deactivation by the formation of β -Ni(OH)₂ or NiO (adapted from Ref. [22]). (c) Electrical circuits, and electrochemical reactions at the cell level in alkaline media. (d) Effects of changes in the electrode microstructure on the OER potential. The electrodes were tested in a 1 M KOH solution in a flow-through single cell equipped with a reference electrode to measure the anode potential. Three Ni porous electrodes with different surface areas and porosities were tested (ordered by decreasing surface area and increasing bubble removal capability): nanowire felt (Ni NW), microfiber felt (Ni MF), and Ni foam (adapted from Ref. [16]). (e) Comparison of the best-performing electrode in Ref. [16] with the state-of-the-art of water electrolysis. Square red symbols represent the maximum current densities for alkaline electrolysis reported in the literature, while the red triangles represent the results for membraneless flow-through alkaline electrolyzers. The shaded areas for AEL and PEM correspond to operating industrial conditions (adapted from Ref. [16]). (f) Innovative cell design proposed by Hodges *et al.*, 2022 [26] where the electrolyte is feed by capillarity (achieved with a polyether sulfone (PES) separator), and bubble formation is avoided. (g) Comparison of the capillary-fed performance at different configurations with standard cells using Zirfon™ as separator (NiFeOOH as anode and Pt/C as cathode). (h) Comparison of the capillary-fed cell performance with commercial alkaline and PEM cells. (i) Sources of improved performance in the capillary-fed cell at 1 A·cm⁻² (f, g, h, and i adapted from Ref. [26]).

optimal pore size related to the balance between increasing surface area and decreasing bubble removal [16]. They have also shown that different types of electrodes with different structures and porosity can impact reducing the ohmic loss stemmed from the bubble formation and accumulation [16]. Modifications

on the separators can also improve their performance at high current densities. Recently, the substitution of porous diaphragms with ion-solvating membranes have resulted in current densities $>1.7 \text{ A} \cdot \text{cm}^{-2}$ at 1.8 V combined with a low hydrogen permeability in cells based on Raney Ni-type electrodes immersed in

Recent catalyst developments.						
Chemistry	Microstructure	Experimental conditions	Overpotential (mV) @ i ($\text{mA}\cdot\text{cm}^{-2}$)	Stability test duration	Research focus	Ref.
Anodes Ru–NiFe	Ru on NiFe defect-rich LDH nanosheets supported on 3D skeleton of Ni foam	O_2 saturated 1 M KOH at 25 °C	220 @ 100	100 h at $100 \text{ mA}\cdot\text{cm}^{-2}$	Increase catalytic activity by structural and chemical changes	[29]
$\text{Ni}_{0.8}\text{Fe}_{0.2}$–AHNA	Ultrathin (1–5 nm) and amorphous-anchored NiFe alloy nanowire (core) array	1 M KOH at 25 °C	248 @ 500	120 h	Increase catalytic activity by microstructural changes	[30]
Pt–Ni(OH)₂@NM	Nanowire/nanosheet composite structure	1 M KOH	269 @ 100	600 h	Increase catalytic activity by chemical changes	[31]
NiFe–HyOx/Stainless Steel (SS)	SS-anode covered by 850 nm-thick NiFe-hydroxide/oxide-catalyst-layer (comprised of particles-like and nano-scale-pores (3–5 nm in diameter))	7 M KOH at 20 °C.	270 @ 100	20,000 potential cycles of 0.5 and 1.8 V	Overpotential and stability comparison between Ni to NiFe–HyOx/SS anodes	[20]
Mo–NiO@NiFe LDH	NiFe-layered double hydroxide (LDH) nanosheets attached on Mo–NiO microrods	6.7 M KOH at 80 °C	253 @ 1000	1200 h	Increase stability and catalytic activity by microstructural changes	[32]
Ni + Co	Ni wire covered with electrochemically deposited Co nanosheets (ns)	1 M KOH at 80 °C + Co-ns dispersion	360 @ 100	40,000 cycles (0.5–1.8 V vs RHE)	Increase stability under fluctuating loads by chemical design changes	[21]
$\text{CuCe}_{0.5}\text{Co}_{1.5}\text{O}_x$	Ce-substituted spinel nanoparticles, phase-transfer coprecipitation synthesis method	1 M KOH	294 @ 10	5000 accelerated cyclic voltammetry at $50 \text{ mV}\cdot\text{s}^{-1}$	Investigate Transition-metal oxides as OER catalysts	[33]
Fe–Ni–Co	Polished metallic surface	1 M NaOH	310 @ 10	50 h	Increase catalytic activity by chemical changes	[34]
Ru–NiFe	Ru on NiFe defect-rich LDH nanosheets supported on 3D skeleton of Ni foam	N_2 saturated 1 M KOH at 25 °C	61 @ 100	100 h at $100 \text{ mA}\cdot\text{cm}^{-2}$	Changes in the local coordination environments of Ru sites to increase performance	[29]
Pt–Ni(OH)₂@NM	Nanowire/nanosheet composite structure	1 M KOH	68 @ 100	600 h	Use PT to improve nickel wire mesh catalytic activity for a low cost and efficient catalyst development	[31]
$\text{Ni}_3\text{N}/\text{Co}_2\text{N}$	Macro porous network structure	1M KOH	70 @ 100	100 h	Increase catalytic activity by interface engineering	[35]
Ni based cathode alloyed with Mo	Nanostructured (periodic surface structures (LIPSS) Ni mesh by femto-laser treatment	7.3 M KOH at 80 °C	135 @ 100	3 days	Increase catalytic activity by changes in the chemistry and microstructure of the electrode	[36]
Fe-13.37Co-11.37Ni-3.21Cr-1.24Mo-0.22C	3D printed martensitic steel with porous and lamellar structures	1 M KOH at RT	800 @ 500	140 h at $570 \text{ mA}\cdot\text{cm}^{-2}$ 450 h at $500 \text{ mA}\cdot\text{cm}^{-2}$	Increase catalytic activity and reduce cost by employing binder free steel electrodes with high active area	[37]

aqueous 24% KOH at 80 °C [27,28]. The stability of such membranes have been improved thanks to a new membrane reinforcement, reaching 1000 h of operation without cell failure [27].

System-level upstream and downstream integration

In the following sections, possible upstream and downstream process integration are presented, including its complexities and recent advances. Figure 2 presents a summary of the integration at a system level.

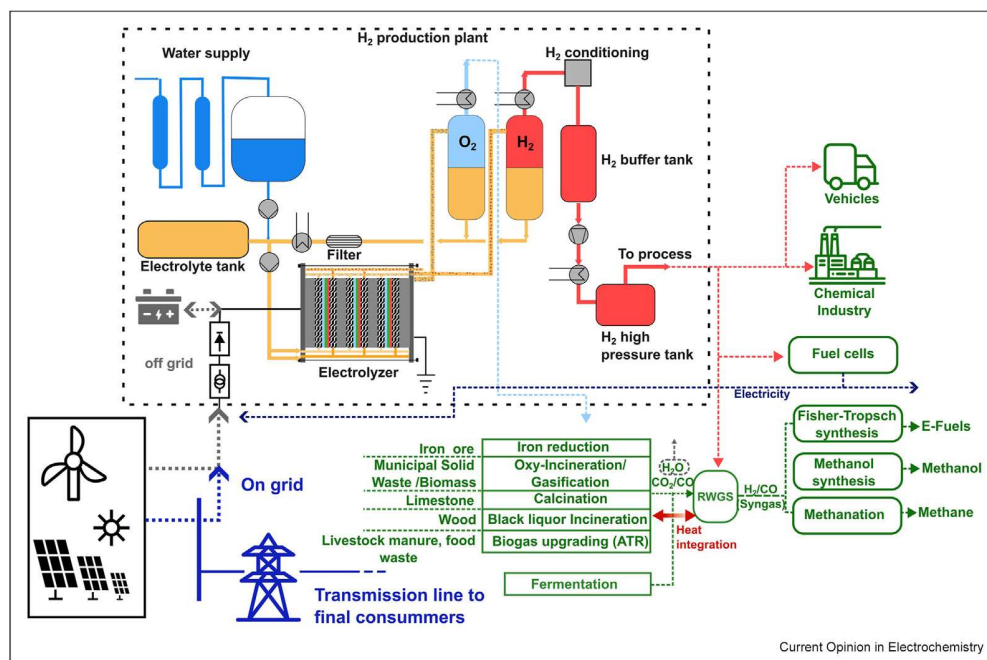
Transient operation and upstream integration including both experimental and simulation

The integration of RES with hydrogen production can be achieved by implementing on-grid (RES and electrolyzer connected to the grid, blue in Figure 2) or off-grid systems (RES and electrolyzer isolated from the grid, gray in Figure 2). The system integration should consider the characteristics of the RES and optimize the topology of the electrical coupling [38,40], the power allocation strategies [41,42] and the operation of the hydrogen plant. In this review, we will focus on the advances related to the aspect of operation.

The inherent RES fluctuation subjects the connected AWE to fast load changes and a wide load range [43]. The

gas impurity is one of the limiting factors in the dynamic operation of AWEs, since it provokes safety shutdowns to prevent the formation of explosive H₂/O₂ mixtures, affecting the stability and power quality of the electrical system [40] and limiting the minimum partial load to 10%–40% of the AWE's nominal power [13,15]. In particular, the lower current densities associated with the partial load will reduce the electrolyzer gas production and consequently reduce the dilution of the impurities, which can result in an unsafe impurity concentration level [4]. The main source of impurity in an AWE is the gas contained in the electrolyte recirculation stream when the system is operated in mixed cycle mode to avoid concentration gradients between the anode and cathode sides [44]. Increasing electrolyte concentration decreases the gas impurity by decreasing gas solubility and diffusion, the upper limit being given by the maximum conductivity achieved by concentrations of 32.5 wt% [44]. Modifications of system operation such as higher temperature operation could improve gas purity in mixed-mode electrolyte circulation by reducing the solubility of the gases and increasing the bubble rise in the electrolyte/gas separators, which is beneficial for the electrolyte/gas separator's efficiency. The operation of a separate electrolyte circuit also allows to obtain safety levels of H₂ in O₂ at current densities as low as 0.05 A·cm⁻² and pressures of 20 bar [45]. However,

Figure 2



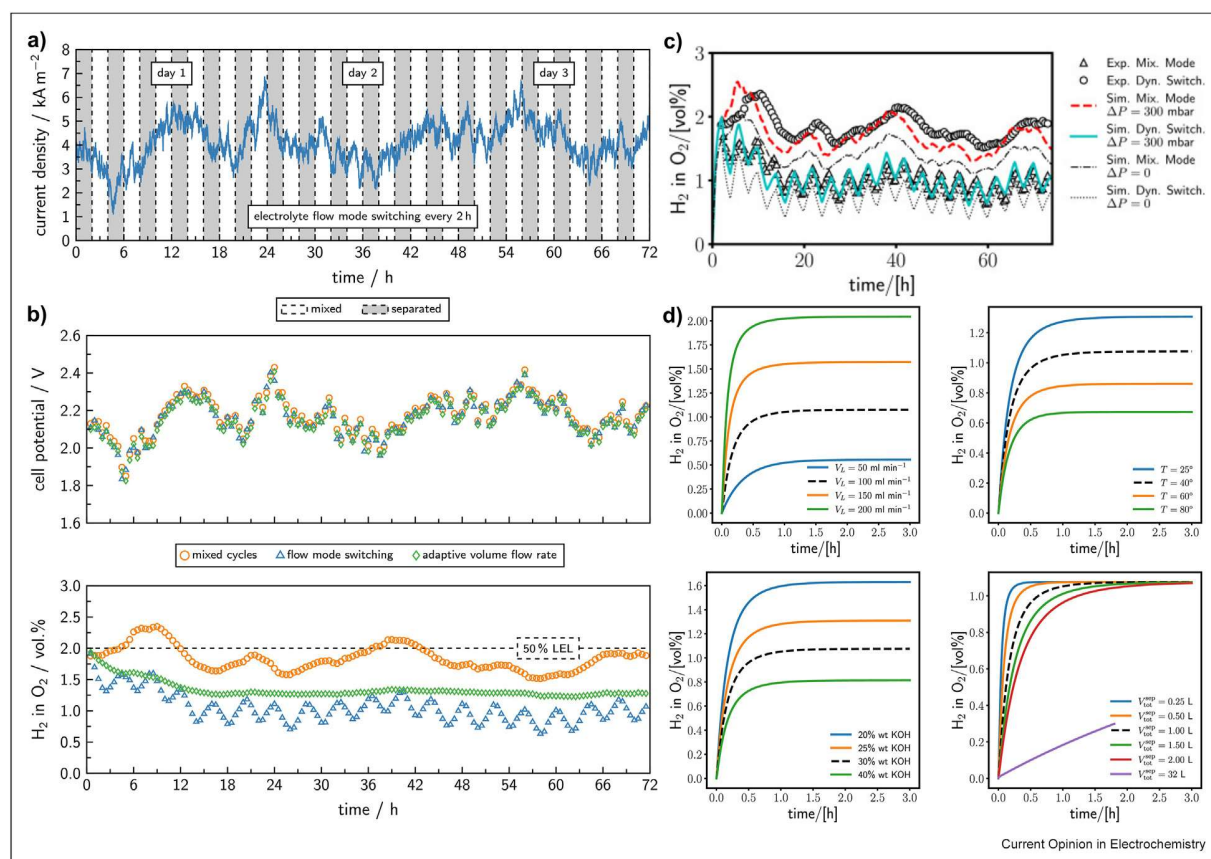
H₂ plant integration into upstream and downstream processes. The boundaries of the H₂ – plant are marked with a dotted black rectangle and include: water purification system, electrolyte circuit (tank, pumps, filter), electrolyzer stack, electrolyte/gas separators (for electrolyte/H₂ separation and electrolyte/O₂ separation before the electrolyte is recirculated into the stack) and final H₂ conditioning and pressurization system. The plant can be integrated into the electricity grid (on-grid) or isolated (off-grid). Battery banks and fuel cells can be installed to balance the system. The produced H₂ can be fed to power to gas and power to liquid process, to the chemical industry or transport sector. Image created based on [14,38,39].

temporary mixing of the electrolyte is required to keep the concentration constant. An alternative is the dynamic operation of the electrolytic cycles, which can be achieved by flexible alternation between mixed and separated modes [44]. The operation with adapted electrolyte flow rate depending on the current density was shown to reduce the gas impurity (to a maximum of 1.3% compared to 2.4% of H₂ in O₂ in standard mode) and does not affect the cell voltage when operating a single cell under simulated wind power profiles, as depicted in Figure 3(a,b) [43]. Nevertheless, the minimum flow rate for each system should be defined, considering cooling requirements [43] and its effect on the bubble growth rate inside the cell [44]. Simulations have been performed to study the combined effect of dynamic loads and different electrolyte circulation modes (Figure 3(c,d)), showing that the dynamic response in terms of impurity accumulation depends on the number

of cells and the gas volume of the system [46]. An impressive impurity level below 1 vol% was achieved by designing multiinput multioutput optimal controllers to minimize the pressure difference between the anode and cathode sides in pressurized electrolyzers, under constant pressure separated electrolyte circulation mode, and fluctuating current emulating RES [47].

The fast load changes associated with RES generate potential and temperature variations, resulting in decreased performance and accelerated cell degradation [48]. Electrolyzers coupled to wind power showed a temperature variation of 8°C when using a simple proportional integral differential (PID) control, which limits the temperature set point to 65°C and leads to low operation performance [49]. Recently, temperature controllers based on transient thermal modeling were shown to be fundamental to mitigate the impact of

Figure 3



Gas purity control in a single-cell electrolyzer. (a) Simulated wind profiles utilized to test different control strategies. The duration of each electrolyte circulation mode is shown for the case of switching from separated to mixed circulation [43]. (b) Variation of the cell voltage and gas purity under the simulated wind profile and different electrolyte circulation strategies (mixed mode and fixed volume flow, mixed mode, and variable volume flow, and separated electrolyte circulation) [43]. (c) The same data [43] was used to validate a model in Ref. [46], where mixed and alternated circulation modes were simulated. The importance of considering additional gas crossover mechanism caused by a differential pressure between cathode and anode sides was highlighted for near perfect agreement between the model and the experimental results. (d) Performing single cell model sensitivity analysis under mixed circulation mode to investigate the dynamic response of the system by varying operation conditions: electrolyte volume flow, temperature, electrolyte concentration, and gas separator volume [46]. Figure adapted from Refs. [43,46].

external disturbances and increase the system efficiency by increasing the temperature set point [50]. Dynamic energy and mass balances could be combined with electrochemical and thermal models to perform sensitivity analyses of changing temperature, mass fraction of KOH in the electrolyte solution and the lye flow rate [51–54], and to develop efficient control strategies.

Researchers have observed that designing a suitable controller for the dynamic operating conditions of AWE systems has been shown to be a powerful tool. It was shown that implementing a multistate transition model (production, stand-by, and off states) led to investigate temperature variations and hydrogen-produced properties, as well as increasing daily revenues [52].

Downstream integration

Figure 2 shows several of the utilization paths of green H₂. Particularly for the mobility sector, chemically stored H₂ in the form of synthetic liquid fuels is considered to be safer and easier to handle than pure H₂. Synthetic liquid fuel production using atmospheric H₂ supply from AWE was simulated and resulted in an energy efficiency of 63.4%. It could be enhanced to 71.5% when system thermal integration was considered [55]. However, for the mobility sector, the overall energy efficiencies reach values close to 21% for E-fuels utilization, while for fuel-cell electric vehicles, this can reach to 31% [53].

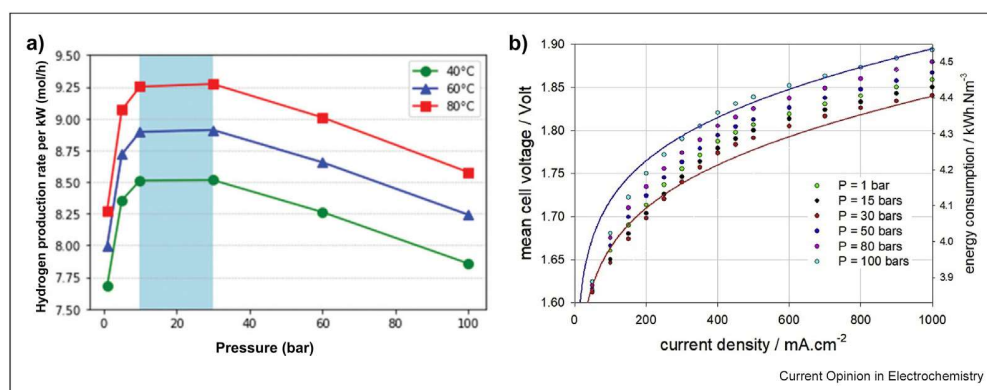
High pressures are generally required for downstream integrations, 200–500 bar for hydrogen transport [56], 700 bar for mobility [56], 20–30 bar for reverse water gas shift (RWGS) units and Fischer-Tropsch reactors [57,58]. Figure 4(a) shows that the AWE system efficiency, calculated by including the energy consumed to compress hydrogen up to 200 bar, reaches a maximum value in the range of 10–30 bars in the stack. However,

further increasing on the stack pressure lead to a decrease of system efficiency. It is also reported that the optimum stack pressure operation could increase with the increase of the final hydrogen compression pressure [59]. It has been reported that incorporating pressurized electrolyzers will contribute to decrease the system cost by reducing the energy consumed for compression [56]. However, pressurization impacts electrolysis energy consumption (as depicted in Figure 4(b)) [60] and increases hydrogen crossover [61]. Moreover, the combination of low current densities with high pressure was shown to exacerbate the unfavorable conditions regarding the gas purity [62]. Strategies using separated electrolyte mode and advanced controllers that minimize the gas crossover could pave the way for the operation at higher pressure under fluctuating loads [47]. New materials have been tested in short stacks reaching up to 100 bar [63], but still more advances are necessary to improve durability and gas purity in a wider load range.

Conclusion

The most recent advances in AWE cell, stack, and system levels were reviewed in this short article. Developing new cell/stack components and cell designs with a high bubble removal rate was shown to be a promising strategy to increase efficiency at high current densities. Furthermore, the need of advanced system operational strategies under transient states for upstream integration and the high pressures required for downstream processes were emphasized. The implementation of advanced control systems for the power supply system's temperature and pressure, and using adaptive electrolyte flow rate and concentration could decrease AWE's minimum load, improve its dynamic response when combined with RES, and increase system efficiency. The downstream integration of highly

Figure 4



(a) Effect of pressure on the hydrogen production efficiency when the energy required for compression up to 200 bar is considered. Results obtained by simulations for an AWE system operating at $0.4 \text{ A} \cdot \text{cm}^{-2}$ [59]. (b) Mean cell voltage variation and energy consumption measured in a 5 cells short stack using optimized electrodes and separators. The experimental results showed an increase in the cell voltage at increasing pressures above 30 bar [63].

pressurized AWE to produce hydrogen, could also increase the overall system efficiency and decrease the system cost by reducing the energy consumed for compression.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data were used for the research described in the article.

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- * of special interest
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A rare high-pressure, up to 100 bar, AWE short-stack of 0.5 kW was tested in this study. Investigations showed that improved and optimized electrodes with a porous catalytic structure and a proper designed diaphragm should be opted for in order to deal with such a pressure. The authors stated that durability reached 1000 h with a limited degradation rate.