



# Overview: Current trends in green electrochemical energy conversion and storage

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Nowadays, hydrogen technologies like fuel cells (FC) and electrolyzers, as well as rechargeable batteries (RBs) are receiving much attention at the top world economies, with public funding and private investments of multi-billion Euros over the next 10 years. Along with these technologies, electrochemical capacitors (ECs) are expanding rapidly in the energy storage market. Electrolyzers, RBs, FCs and ECs are electrochemical energy conversion and storage devices offering environmental and sustainable advantages over fossil fuel-based system. This overview discusses current trends in these electrochemical systems. It also examines recent advances on the CO<sub>2</sub> reduction reaction, which has gained attention because of the capability of producing value-added chemical products at high efficiencies from the main greenhouse gas in the USA and the European Union. It is expected that these green systems will play a key role in the imminent implementation of a global sustainable energy scenario.

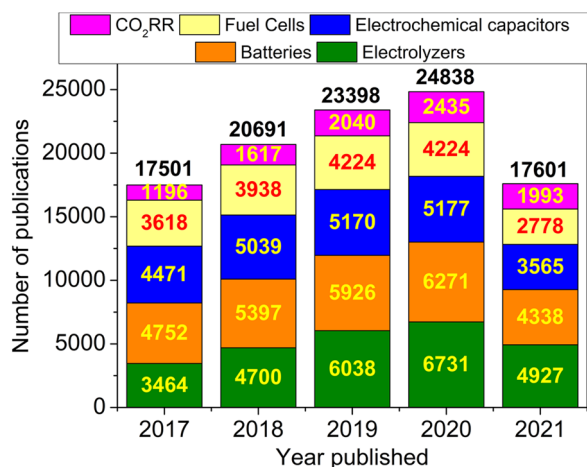
## Introduction

Electrochemical energy conversion and storage devices, and their individual electrode reactions, are highly relevant, green topics worldwide. Electrolyzers, RBs, low temperature fuel cells (FCs), ECs, and the electrocatalytic CO<sub>2</sub>RR are among the subjects of interest, aiming to reach a sustainable energy development scenario and reducing the dependence on fossil fuels.

The increasing interest on these issues is demonstrated in Fig. 1 by the number of publications over the 2017–2021 period based on a Web of Science search of topics shown in Table S1. The total publications per year are shown at the top of each column. A continuous rise in publications can be observed over the years. Electrolyzers, RBs, ECs, and FCs show a growing research interest even though FCs publications are constant in the 2019–2020 period. Meanwhile, the CO<sub>2</sub>RR research has grown over the past few years.

The growing interest is motivated by the advantages offered by the electrochemical systems and processes compared to fossil fuel-based devices, among them: sustainable operation, high efficiency or high cyclability, and the clean production of chemicals of industrial interest from greenhouse gas (GHG) CO<sub>2</sub> [1–3]. FCs, RBs, and ECs offer advantages from a sustainable point of view over conventional systems in a wide variety of energy conversion and storage applications, including: (i) stand-alone and portable systems; (ii) residential appliances; (iii) electric vehicles; (iv) aerospace programs [4–6]. For instance, considering their application in the automotive industry, it has been reported that CO<sub>2</sub> emissions can be reduced by 30% following the production of 20 million electric, plug-in hybrid electric, and fuel cell vehicles by 2050 [7].

Meanwhile, water electrolysis produces high-purity hydrogen and oxygen gases [8]. Adding-up environmental

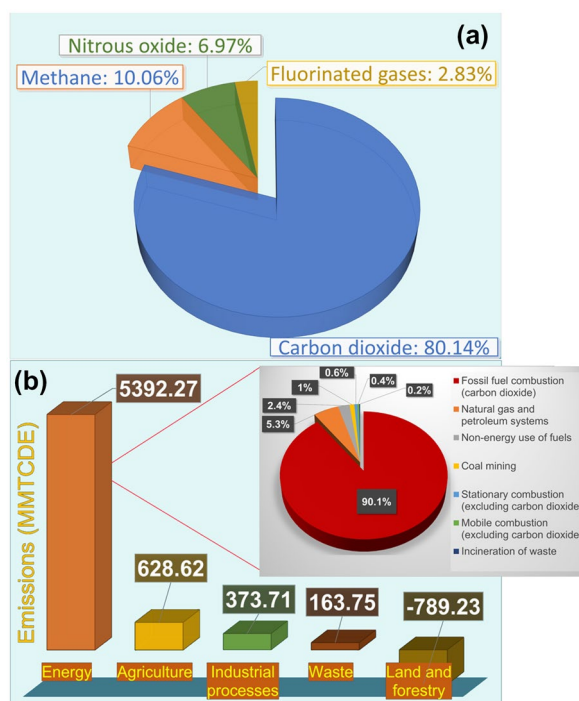


**Figure 1:** Number of publications on electrolyzers, rechargeable batteries, low temperature fuel cells, electrochemical capacitors, and their electrode reactions, as well as the CO<sub>2</sub>RR. Source: Web of Science, as of September 7, 2021.

aspects, green hydrogen and oxygen can be produced by integrating electrolyzers and renewable energy systems, such as solar and wind, at competitive costs under given operating conditions. In a recent study, researchers report the economic profitability of 100 kW to 10 MW electrolyzers in two scenarios with self-production of oxygen, in models that consider hydrogen selling at 10 € kg<sup>-1</sup> [9]. In another study, it has been reported that the decarbonization of hydrogen production can be implemented at a regional level in the European Union. It is reported that, after covering the yearly demand of electric energy, 88 European regions have excess potential electricity from renewables to produce 9.75 Mt of green hydrogen. For such task, 290 TWh of electricity would be required [10].

The production and use of green hydrogen and oxygen limit the emission of CO<sub>2</sub> and other GHGs. Additionally, CO<sub>2</sub> can be electrochemically transformed into value-added products such as CO, HCOOH, C<sub>2</sub>H<sub>4</sub>, C<sub>2</sub>H<sub>5</sub>OH, and C<sub>3</sub>H<sub>8</sub>O, among others via the CO<sub>2</sub>RR [3]. Electrochemical CO<sub>2</sub>RR has gained importance because of its high Faradaic efficiency, especially in the production of CO and HCOOH [3]. Transforming CO<sub>2</sub> into value-added products is of vital relevance, since according to the inventory of the Environmental Protection Agency (EPA), this is the main GHG in the USA [11].

Per the data available in 2019, CO<sub>2</sub> accounts for 80.14% of the GHGs emitted (in million metric tons of carbon dioxide equivalent, MMTCDE), as seen in Fig. 2a. The energy sector is the main source, with 5392.27 MMTCDE, followed by agriculture, industrial processes, and waste (Fig. 2b). Breaking down

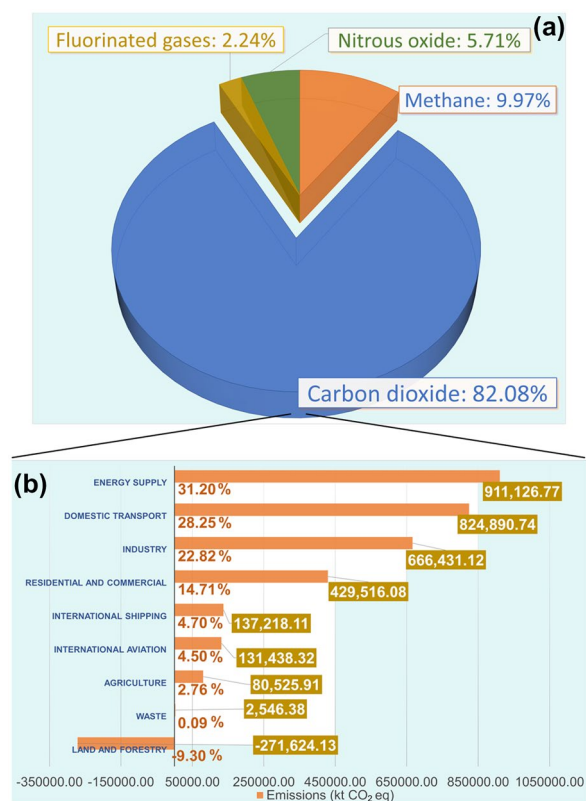


**Figure 2:** Emissions of GHGs in the USA. (a) Percentage per gas and (b) emissions by category. Inset in (b): Breakdown of emissions in the energy category. Note: Land and forestry is short for Land use, Land Use-Change, and Forestry. Source: EPA's Greenhouse Gas Inventory Data Explorer [11].

the energy sector, fossil fuel combustion contributes 90.1% of CO<sub>2</sub> emissions, as can be seen in the inset of Fig. 2b. Meanwhile, land and forestry (in fact: Land use, Land Use-Change, and Forestry), which act as sinks, have a negative value.

The same trend of GHG emissions in the EU has been reported by the European Environment Agency (EEA, geographic entry EU-27) [12]. As seen in Fig. 3a, CO<sub>2</sub> accounts for 82.08% of the total kilo tonnes of CO<sub>2</sub> equivalent (kt CO<sub>2</sub> eq) of GHG emitted, as of 2019. A breakdown of these emissions show that the energy supply sector contributes with 31.20% of CO<sub>2</sub> emissions (911,126.77 kt CO<sub>2</sub> eq) and domestic transport in second place with 28.25% (Fig. 3b) [12].

The figures presented here show the relevance of advancing the massive implementation of electrochemical energy conversion and storage devices, aiming for a sustainable global energy scenario. Each of the devices already mentioned have advantages, but also technical drawbacks, that need to be addressed by the scientific community. In the following sections, the most recent advances regarding these systems are presented. Moreover, this focus issue presents the most recent works of several international Laboratories on the topics of electrolyzers, RBs, low temperature FCs, ECs (and their electrode reactions), and the CO<sub>2</sub>RR.



**Figure 3:** Emissions of GHGs in the EU. (a) Percentage per gas and (b) breakdown of CO<sub>2</sub> emissions by category. Land and forestry is short for Land use, Land Use-Change, and Forestry. Source: EEA's Greenhouse Gases—Data Viewer [12].

### Low temperature electrolyzers: green hydrogen and oxygen from water splitting

Proton exchange and anion exchange membrane electrolyzers (PEME and AEME, respectively) are all-solid, modular electrochemical devices for water splitting. These technologies aim to compete with the more mature, conventional alkaline electrolyzers (AE) that typically use aqueous KOH as electrolyte. PEME and AEME systems have the capacity of producing high-purity hydrogen and oxygen (> 99.999% and > 99.99, respectively) from the electrolysis of H<sub>2</sub>O molecules, with a high energy efficiency (80–90%) [13, 14]. Meanwhile, the gas purity from AE is lower (> 99.5) [14]. The lifetime of PEME systems is around 20,000–60,000 h, less than that of AE (~ 90,000 h) [14, 15]. It has been reported that the lifetime of AEME systems is < 3000 h [16].

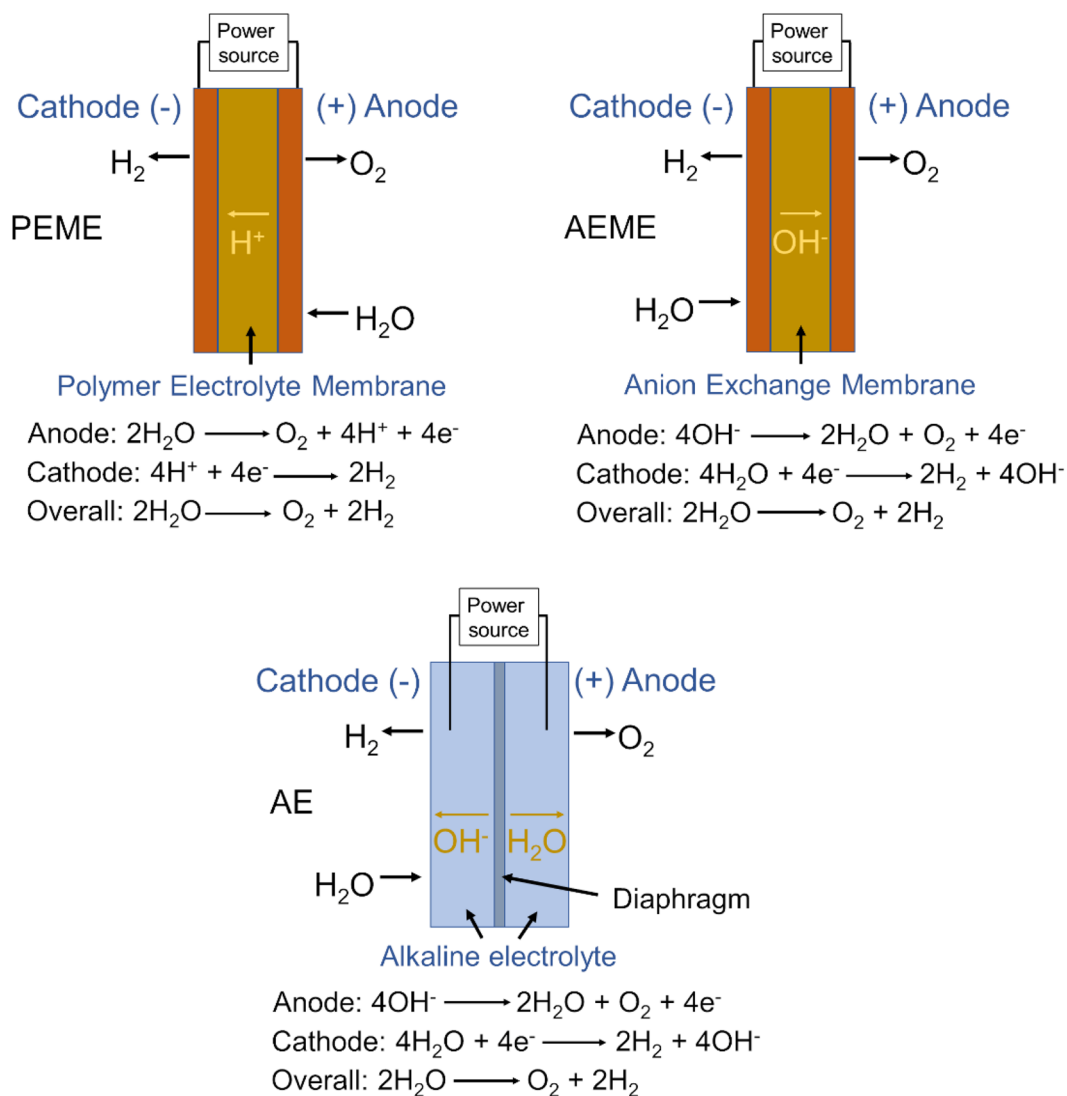
PEME and AEME share some similarities in their configuration, distinguishing from AE in the use of a solid polymer electrolyte. Their temperature of operation typically ranges from 50 to 90 °C. At the anode, the hydrogen evolution reaction (HER) takes place, while the oxygen evolution reaction (OER) proceeds at the anode. Schemes of PEME, AEME, and

AE, including their electrode and overall reactions, are shown in Fig. 4.

Alongside the production of high purity H<sub>2</sub> and O<sub>2</sub>, which by itself is of scientific and technological interest, electrolyzers can be powered with renewable energy sources such as solar and wind. Thereby, the green hydrogen concept has gained international relevance because of its positive impact in the decarbonization of power supply, seeking to achieve a carbon neutral energy scenario. The production of green hydrogen from water electrolysis powered by renewables has the potential of reducing the annual carbon emissions by 10.2 Gt, assuming the RCP8.5 scenario from the International Panel on Climate Change as Ref. [17]. Moreover, the consumption of water for green hydrogen production has been reported to be significantly low, i.e., annual consumption of 1.5 ppm considering freshwater or 30 ppb regarding saltwater on Earth, figures that are smaller compared to those of other sectors based on fossil fuels [17]. Green hydrogen can replace the so-called grey hydrogen, which currently is mainly produced (96%) from fossil resources, emitting 830 million tons of CO<sub>2</sub> to the atmosphere per year [18]. Overall, it is estimated that global electrolyzer capacity will increase to 1.5 GW in 2023 [19].

Green hydrogen production using AE has been reported. A 250 kW AE (50 m<sup>3</sup> h<sup>-1</sup> H<sub>2</sub> production) has been shown to produce 99.5% purity H<sub>2</sub> with a DC energy efficiency in the 73.1–65.0% LHV range, under wind power supply simulations for more than 4000 h [20]. In an analogous sustainable fashion, production of green hydrogen in South America using waste energy from hydroelectric power plants has been demonstrated. Paraguay shows the highest production and storage profit (0.2234 US\$ kg<sup>-1</sup>), compared to Argentina and Uruguay [21].

Production of green hydrogen using PEME has also been explored. A system integrated by solar photovoltaic as the power source, a PEME (99.9999% H<sub>2</sub> purity), lead batteries, and metal hydride tanks as H<sub>2</sub> storage unit has been proposed. The PEME efficiency oscillates between 50 and 70%, while production of H<sub>2</sub> by the system ranges from 65 to 85%. The average global efficiency of the system is ca. 19%. The green hydrogen production rate at the system has been reported as 115 L day<sup>-1</sup>, with a cost of 1.09 € m<sup>-3</sup> [22]. Elsewhere, a comparison of PEME and AE technologies indicates a higher performance of the latter regarding hydrogen production, due to its maturity. Nevertheless, PEME has shown about 13–15% higher efficiency. According to the report, to produce 180 kg day<sup>-1</sup> H<sub>2</sub>, a PEME system requires ca. 10 MWh per day, achieving a system efficiency of nearly 60%. Regarding thermal energy management, the PEME required less cooling energy [23]. Nevertheless, another study indicates a hydrogen production efficiency of 78–84% from a kW-scale AE [24]. Similarly, a recent work reports a stable performance of an AEME over 10,000 h testing, degrading by 0.15 μV cycle<sup>-1</sup> after 11,000 cycles [25].

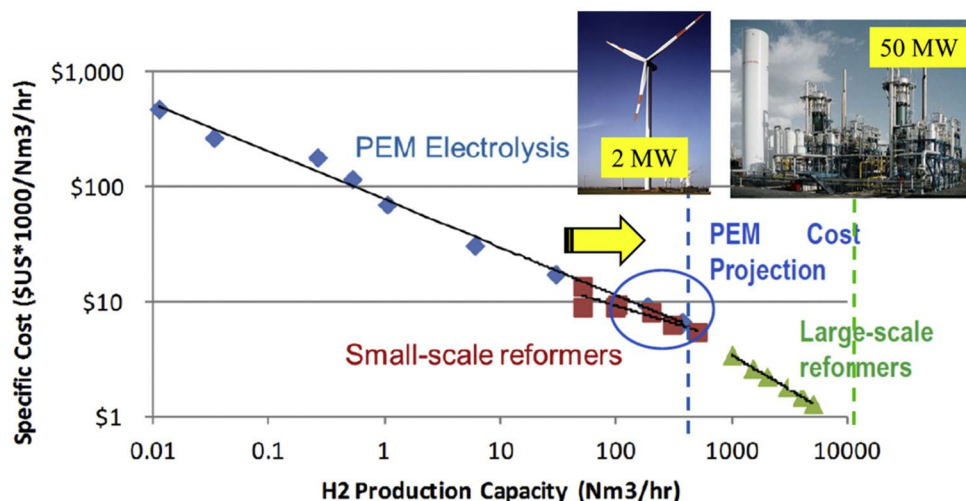


**Figure 4:** Schemes and electrode reactions of PEME, AEME, and AE.

In order to compete with steam-methane reforming (SRM), costs of hydrogen generation from water electrolysis must decrease while sustaining a high production capacity. Moreover, the unit size of electrolyzers has to be scalable to the point to reach the requirements due to the dimensions of renewable energy systems. In other words, electrolyzers for the production of green hydrogen with solar and wind need to move into the MW-size. As reported earlier, scaling-up the electrolyzers can have a positive impact in reducing the capital expenditure (CAPEX), putting them in a competitive position against SRM units [26]. Figure 5 shows a specific cost vs. hydrogen production capacity plot. It is projected that by reaching the MW-size for interconnection with off-shore wind turbines, the CAPEX of electrolyzers will have the potential to compete with small-scale SMR systems [26].

### Rechargeable batteries (RBs): Versatile high energy density storage systems

RBs are an essential technology for energy storage in the current lifestyle. Furthermore, RBs in conjunction with hydrogen technologies presently play a key role in achieving a carbon neutral economy, according to the 2050 scenario proposed by the EU [27]. The main contribution of RBs to reach this goal is their use in the transportation (electric vehicles) and energy (renewable energy power storage) sectors. There are several types of RBs; however, the Li-ion batteries (LIBs) industry has grown rapidly, from a global manufacturing capacity of 103.7 GWh to 273 GWh in the 2017–2021 period [28]. LIBs are of interest due to their outstanding advantages over other technologies, such as high energy density ( $250 \text{ Wh kg}^{-1}$ ) [29], life cycle up to

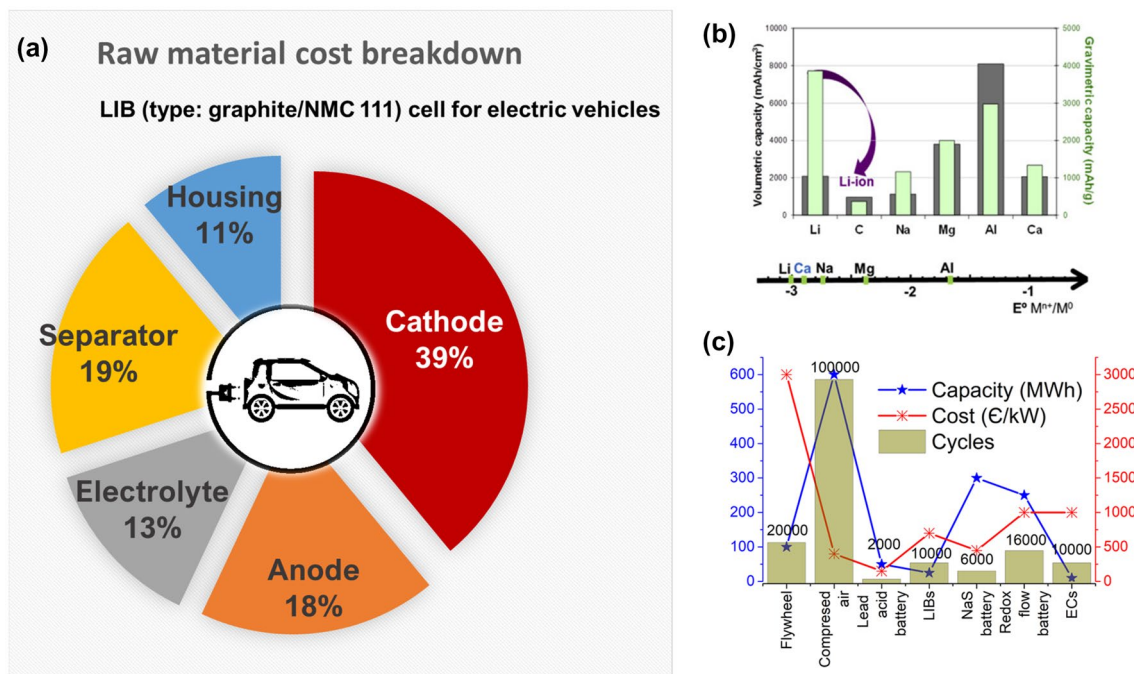


**Figure 5:** Scaling-up electrolyzers for green hydrogen production can lead to significant cost reductions. In the plot, blue diamonds in the 0.01–100 Nm<sup>3</sup> h<sup>-1</sup> range represent real data for PEM electrolyzers, while those in the 100 to 1000 Nm<sup>3</sup> h<sup>-1</sup> interval are cost projections. Red squares and green triangles represent real data of SMR units of small-scale (on-site) and large-scale (centralized) capacity, respectively. Reprinted with permission from [26]. Copyright (2020) Elsevier.

1000 cycles (at 80% discharge), high cell voltage (3.3 to 3.8 V), minimal maintenance, and low toxicity [30].

The main drawback of LIBs is their high cost ( $\approx 200$  to  $400$  \$ kWh<sup>-1</sup>). In fact, many production plants are subsidized [31]. Raw materials constitute the main cost in the production process of conventional LIBs ( $\approx 58\%$  of the total cost),

the cathode being usually the most expensive component. In this regard, materials cost breakdown of a typical graphite/NMC (LiNi<sub>0.33</sub>Mn<sub>0.33</sub>Co<sub>0.33</sub>O<sub>2</sub>) battery is shown in Fig. 6a. As can be seen, the cost of the cathode represents 39% of the total cost of the raw materials, and approximately 24% of the total production cost [32]. Therefore, the development of advanced



**Figure 6:** (a) Raw material cost breakdown of LIBs (type: graphite/NMC 111) used in electric vehicles. (b) Standard reduction potential and volumetric/gravimetric theoretical capacities of multivalent ions used in next generation batteries. Reproduced with permission from [36]. Copyright (2019) Elsevier. (c) Comparison of capacity, cost and cyclability of different technologies used in energy storage for renewables.

lithium-based batteries such as lithium sulfur batteries (LSB), solid state batteries (SSB), and lithium-air batteries (LAB) in terms of reducing costs is very attractive. However, nowadays these technologies are produced on a pilot scale.

Besides cost-efficiency issues, there are concerns about the environmental benefits of large-scale production of LIBs. Recently, it has been reported that the eco-efficiency of manufacturing LIBs (i.e., the simultaneous economic and environmental impacts of developing these technologies) can be as low as 39.5 kg CO<sub>2</sub> eq (kW h)<sup>-1</sup> [28]. At the same time, the cost of advanced lithium-based batteries foresees a reduction to below 90 \$ kW h<sup>-1</sup> by 2050 [33].

Regarding electric vehicle applications, the DOE target by 2022 envisions a range of at least 500 km, at a cost of 125 \$ kW h<sup>-1</sup>. Currently, mature LIBs technologies such as layered LiNi<sub>1-x-y</sub>Mn<sub>y</sub>Co<sub>z</sub>O<sub>2</sub> (NMC), LiMn<sub>2</sub>O<sub>4</sub> (LMO), and LiNi<sub>0.8</sub>Co<sub>0.15</sub>Al<sub>0.05</sub>O<sub>2</sub> (NCA) are used in electric vehicles. However, it is anticipated that by 2035, advanced batteries such as LSB, SSB, and LAB will replace current technologies. Another important issue to consider is the availability of raw materials. For NMC, LMO, and NCA batteries no supply difficulties exist in the short- and long-term periods for materials such as graphite, nickel, copper, and aluminum [34]. Meanwhile, it is anticipated that supply of critical materials such as lithium and cobalt may not cover the global demand by 2035 [35].

Because of the difficulties related to LIBs, less expensive technologies have been developed for the next generation batteries: the sodium-ion batteries (SIBs) and the multivalent-ion batteries (i.e., Mg<sup>2+</sup>, Ca<sup>2+</sup>, Al<sup>3+</sup>), having an estimated energy density of up to 450 Wh kg<sup>-1</sup> [36]. SIBs and multivalent-ion batteries are still in the development stage. Figure 6b shows the standard reduction potential and the theoretical volumetric and gravimetric capacities of some multivalent ions evaluated in a cell with a graphite anode. As can be seen, the Al<sup>3+</sup> ion batteries are very promising in terms of capacity. However, they have two important limitations to overcome: (i) more stable electrolytes to avoid corrosion issues must be developed, and (ii) currently, there are no cathode electrodes that can sustain the 3 e<sup>-</sup> transfer of the Al<sup>3+</sup> reaction during long-term operation [37].

Another important sector is the energy storage in renewable systems. It is estimated that the LIB market will grow from 2 GW in 2017 to 175 GW in 2030. Even though there are several energy storage technologies in stationary systems (i.e., mechanical storage such as flywheel, compressed air, pumped hydro, among others), LIBs are considered the most important among the electrochemical technologies.

Figure 6c shows a comparative scheme of capacity (MWh), cost (€ kW<sup>-1</sup>), and cyclability of different energy storage technologies employed in stationary systems [38]. Compressed air systems show the highest capacity (≤ 600 MWh), with a relatively affordable cost (400 € kW<sup>-1</sup>), along with a high cyclability

(~ 100,000 cycles). However, geographic issues and the environmental impacts associated with the installation of these systems are usually very negative. LIBs have good capacity (0.25–25 MWh), cost range (350–700 € kW<sup>-1</sup>), and cyclability (~ 10,000 cycles), making this technology a fair alternative. The main obstacle of LIBs is their high cost and safety issues related to their operation.

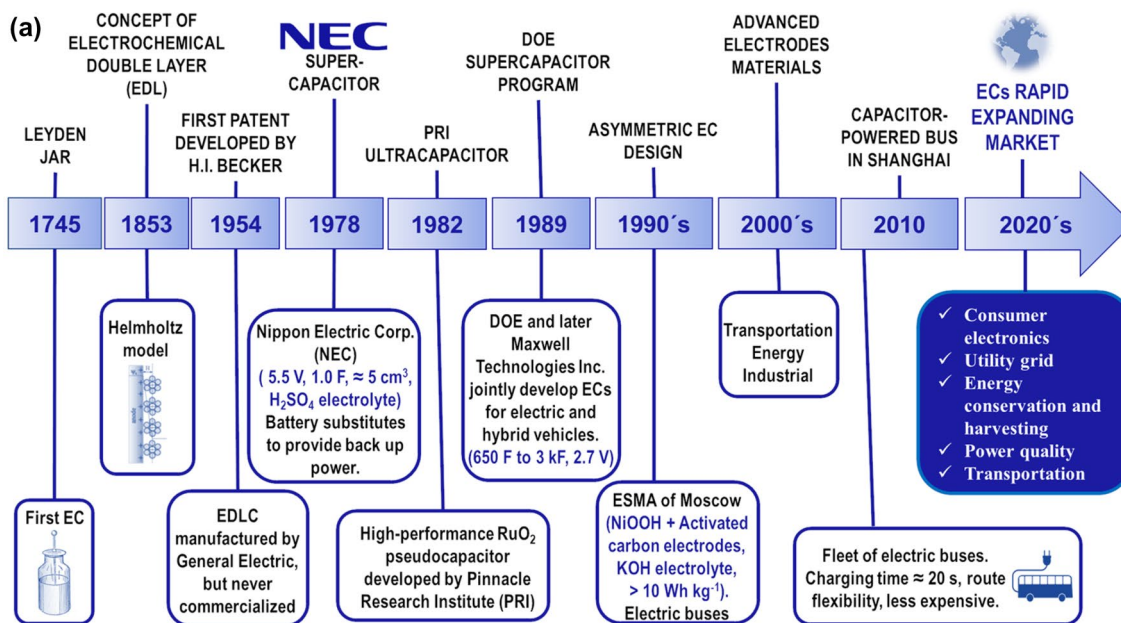
## Electrochemical capacitors (ECs): Rapidly expanding market for green energy storage

ECs are not a new technology. The first EC was developed in 1764 (Leyden Jar), and almost a hundred years later the concept of electrochemical double layer (EDL) was postulated by Helmholtz in 1853. Coincidentally, almost a hundred years later the first EC was patented by General Electric Co. in 1954. Thenceforth, several companies have developed their own ECs for different applications. Figure 7a shows the timeline with the most relevant events in the history of ECs development [39]. Additionally, this figure has information of some importance for manufacturing companies, applications, and characteristics of their ECs.

Currently, the ECs market is expanding rapidly for many applications (Fig. 7a), mainly due to the improvement of its performance over time. ECs are expected to reach a market of US\$1869.05 million by 2026, growing annually by 13.5% over the 2021–2026 period [40]. However, it is necessary to continue improving this technology to overcome the current risks in the market, especially those related to increasing its energy density without compromising its high-power density. In this section, the most important challenges that concern ECs are classified into 4 subsections, addressing the most relevant solutions proposed thus far.

- (i) Development of new/optimized sustainable materials.

Figure 7b shows the components of ECs, which can be classified as active (i.e., electrodes and electrolyte) and passive (i.e., separator, binder, and current collector) [41]. The major contribution related to energy storage performance is from the electrodes. It is from the storage mechanism that the classification of ECs is derived [42, 43]. This subsection focuses on materials developed for advanced asymmetric electrochemical capacitors (AECs), due to their notable advantages and promising performance, such as high energy density and extended operating voltage window [31]. Some parameters reported for AECs can be seen in Fig. 7b. Regarding electrode materials, metal organic frameworks (MOFs) [44] and covalent organic frameworks (COFs) [45] have been identified as potential candidates to increase the energy density and improve ion diffusion. However, the



**(b) Asymmetric Electrochemical Capacitors (AECs)**

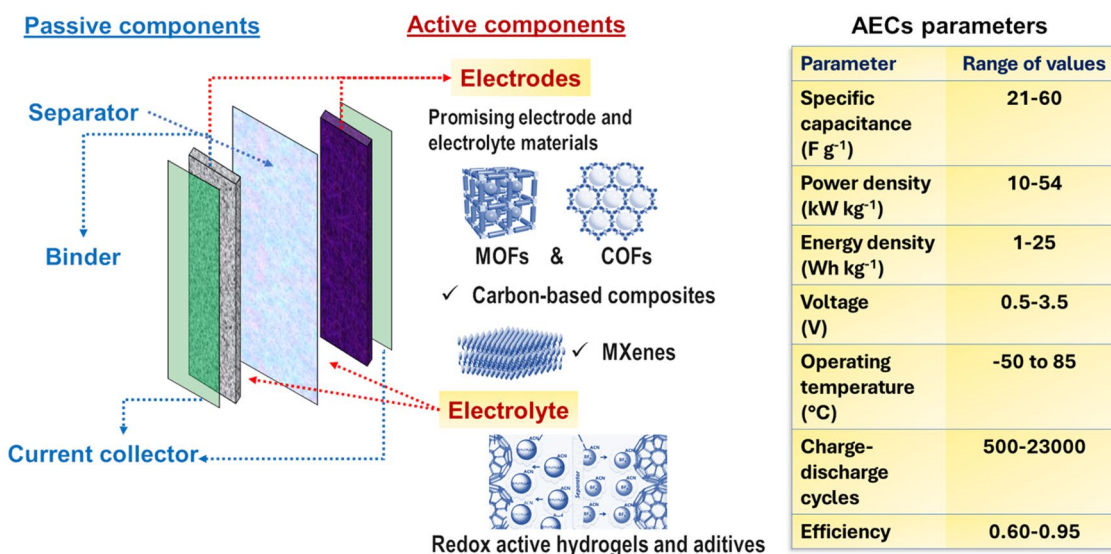


Figure 7: (a) Development timeline of ECs and (b) scheme of an AEC showing its components. Inset in (b): The main parameters of AEC.

main drawback of these materials is their low electrical conductivity, which has led to the study of composites which include carbon-based materials [46].

From 2011 to date the interest in research of a new family of two-dimensional transition metal carbides and carbonitrides named MXenes has significantly increased [47]. MXenes behave like “conductive clays”, attributed to their intrinsically conductive and hydrophilic nature [48]. Other potentially promising materials for AEC electrodes are black phosphorus and tran-

sition metal dichalcogenides [49, 50]. Similarly, the electrolyte has a relevant effect on the overall performance of AECs. Ionic conductivity, toxicity, and thermal and electrochemical stability are key parameters for proper electrolyte selection. On this issue, novel hydrogels redox-active electrolytes are safer, easier to synthesize and have lower cost than organic electrolytes [51]. Another novel approach that significantly enhances the energy density of AECs is adding redox-active species to the electrolyte [52].

- (ii) Implementation of novel tools to fundamental understanding.

Specialists in the field state that it is necessary to develop *in situ operando* techniques that allow a fuller understanding of the complex charge-storage and self-discharge mechanisms at ECs [42, 51]. In addition, it is necessary to couple experimental approaches with theoretical modeling or computational simulation to study more complex electrode/electrolyte interface structures. For example, soft X-ray spectroscopy coupled with theoretical modeling has been recently used for the *in operando* study of ECs electrodes [53].

- (iii) Novel designs for systems innovation.

The fast growth of EC applications demands not only the manufacture of advanced electrodes or electrolyte materials. It also involves innovating in their conventional design. On this matter, novel designs of ECs can play an important role for improving their performance. Some examples of novel designs are: (i) electrochemical flow capacitors, which provide a rapid energy storage and recovery for stationary systems [54]; (ii) multivalent metal ion hybrid ECs (i.e., Zn, Al, Mg, Ca ion ECs) which integrate the advantages of metal ion batteries and ECs with a more sustainable approach [55]; (iii) micro-ECs (i.e., fiber-shaped, plane and three-dimensional) [56]; and (iv) multifunctional ECs such as electrochromic, self-healing, piezoelectric, shape-memory, thermal self-protecting, thermal self-charging and photo self-charging systems [57].

- (iv) Tests for performance evaluation.

A huge limitation for the comparison of performance of ECs developed by different manufacturers, is the lack of standardized criteria for characterization and testing. While some manufacturers have adopted their own procedures, there exist those recommended by the US Department of Energy (DOE), the standards by the Chinese agencies (for example, the QC/T 741-2014), and that of the Standards Institution of Israel (SII NWIP), among others. A detailed comparison of several testing procedures and protocols has been reported recently [58], which includes the measurement of advanced ECs. The range values of various parameters related to AECs are shown in Fig. 7b.

## Low temperature fuel cells: Clean energy conversion at high efficiency

Polymer Electrolyte Membrane Fuel Cells (PEMFC) and Anion Exchange Membrane Fuel Cells (AEMFC) are also all-solid, modular devices for clean, CO<sub>2</sub>-free, electrochemical energy generation. Their temperature of operation is commonly

between 60 and 85 °C [4, 59]. PEMFC can reach an electrical efficiency of 40–80% [4]. The lifetime of a PEMFC is typically longer than that of an AEMFC. The performance of a stationary 70 kW PEMFC stack over a period of 30,000 h has been reported. The best MEAs show an average voltage decay of 2.5 μV h<sup>-1</sup> over 16,000 h [60]. Meanwhile, AEMFCs have demonstrated shorter lifetime. For example, a recent work indicates a lifetime of AEMFC of 550 h [61]. However, longer values have been reported, including 2000 h operation at 75 °C and 600 mA cm<sup>-2</sup>, with a decay of 15.36 μV h<sup>-1</sup> [62].

PEMFCs typically use Nafion<sup>®</sup>, a perfluorinated membrane as the polymer electrolyte. The structure of Nafion<sup>®</sup> is that of polytetrafluoroethylene backbone, with perfluorinated chains having SO<sub>3</sub>H groups in the end [63]. The chemical stability of Nafion<sup>®</sup> is high. It possesses a high proton conductivity (0.152 S cm<sup>-1</sup> at 90 °C), which according to one recent study can increase to 0.211 S cm<sup>-1</sup> using phosphotungstic acid-modified cobalt oxide nanoparticles as filler [64]. Some of the main drawbacks of Nafion<sup>®</sup> are the accelerated dehydration at temperatures around 100 °C, decreasing its conductivity [65], and its cost, reported as high as 1733 US\$ m<sup>-2</sup> for Nafion 117, and ca. 800 US\$ m<sup>-2</sup> for Nafion 212 [66, 67].

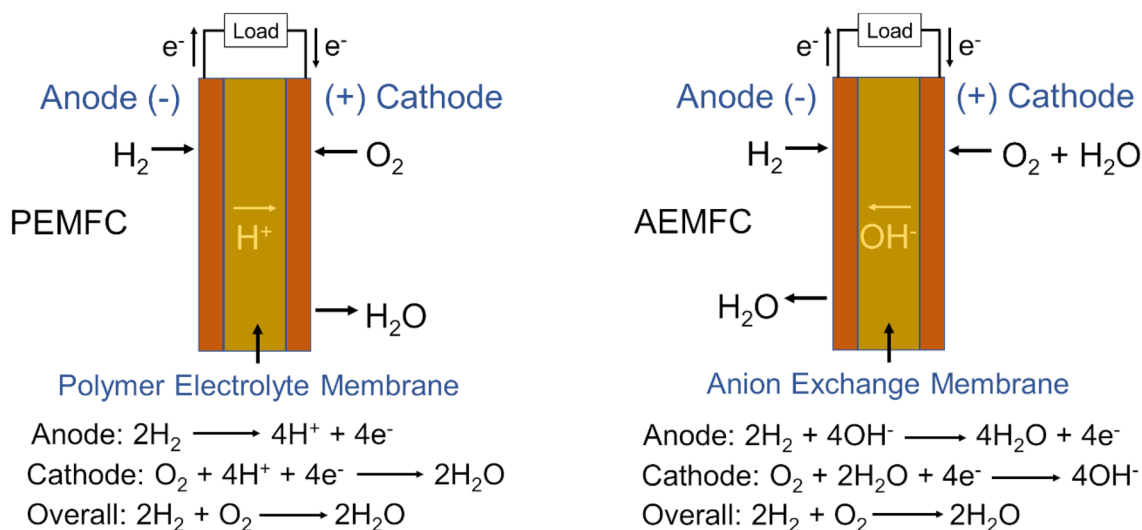
PEMFCs have been investigated for a longer time than AEMFCs, reaching a commercial status, including testing on-field operation for over 10,000 h [68]. Their technological advancement is such that are commonly considered for 1 to 250 kW scale power systems [69]. Even more, several automotive companies commercialize fuel cell electric vehicles based on this technology [69–72]. Nevertheless, it is acknowledged that one of the main issues to overcome for a broader commercialization of PEMFC is their cost, which must be reduced to compete with energy systems based on fossil fuels.

As an example, it has been reported that the cost of fuel cell electric vehicles from major companies lies in the \$58,000–\$62,000 range, with an autonomy within 312–380 miles (502–611 km) [72]. The FC stack power used in these vehicles spans from 90 to 128 kW [69, 71]. It has been reported that the new generation Mirai (Toyota, commercialized in 2020) achieved 4.4 kW L<sup>-1</sup> and 5.4 kW L<sup>-1</sup> stack power density with and without endplates, respectively. Such figures represent an increase of 42 and 54% compared with the previous generation [73].

Figure 8 shows schemes and electrode reactions of PEMFC and AEMFC. At the anode, the hydrogen oxidation reaction (HOR) takes place, while the oxygen reduction reaction (ORR) proceeds at the anode.

AEMFCs have the advantage that non-Pt group metal (PGM) catalysts can be used to promote the HOR and the ORR [74–76], which lowers their cost [77]. However, one disadvantage of AEMFC is the sluggish kinetics of the HOR and the ORR in alkaline media, even using PGM at the electrodes [77].





**Figure 8:** Schemes and electrode reactions of PEMFC and AEMFC.

Another issue with these devices is the polymer electrolyte. Several anion exchange membranes have been tested in AEMFC, having the goal of reaching the performance and commercial status of Nafion® [78]. Some membranes that have been commercialized, and their ion conductivity ( $\text{mS cm}^{-1}$ ) are [79]: (i) Fumasep® FAA3 from Fumatech, 4–7 (Cl) or 40 (OH); (ii) A201 from Tokuyama, 42 (OH); (iii) AEMION™, > 80 or between 15 and 25. The target is developing membranes with ion conductivity > 50  $\text{mS cm}^{-1}$ .

According to a recent report, a majority of experts in the field of PEMFC consider that the cost of PGM catalysts, because of the metal loading, is the main barrier for the reducing the costs of this technology. In second place emerge the cost of the membranes, while third place is for bipolar plates [80]. This ranking may apply for AEMFCs too.

### Electrochemical CO<sub>2</sub> reduction reaction (CO<sub>2</sub>RR): Value-added chemical products

There has been an increase in the global atmospheric concentration of CO<sub>2</sub>, reaching worrying 385–415 ppm values in the past few years, from 280 ppm in 1750 [81, 82]. At the same time, CO<sub>2</sub> is a much-needed carbon source for the nourishing of Life on the planet [83]. Among several synthesis routes, CO<sub>2</sub> can be electrochemically converted to value-added C1 and C2 chemical products, which are of interest for the industry. The CO<sub>2</sub>RR is a complex, multi-electron/proton transfer mechanism, requiring the use of heterogeneous nanocatalysts to improve the reaction kinetics [3, 81, 82].

The Faradaic efficiency is limited by several factors, including the solubility of CO<sub>2</sub> in aqueous electrolytes [84].

Meanwhile, parameters such as selectivity, activity, and stability are related to the nature of the nanocatalyst, its morphology, the electrolyte composition, and the process conditions [85]. Faradaic efficiencies of CO<sub>2</sub> to CO conversion > 95% have been reported [81]. Lower Faradaic efficiencies have been reached for C<sub>2</sub>H<sub>4</sub> (80%) [86], C<sub>2</sub>H<sub>5</sub>OH (52%) [87], and n-C<sub>3</sub>H<sub>7</sub>OH (30%) [88]. Moreover, a Faradaic efficiency of ca. 97% has been reported for HCOOH [89].

Being a relatively new technology, there are not commercially available CO<sub>2</sub>RR systems [85]. According to a techno-economic analysis, the CO<sub>2</sub> reduction resulting in HCOOH and CO may reach a competitive status compared to conventional processes, while other products may not be economically viable, unless environmental and sustainability issues are brought into consideration [85]. Nevertheless, the wide variety of products that can be obtained from the CO<sub>2</sub>RR is quite unique [90] and provides potential commercialization advantages to the technology.

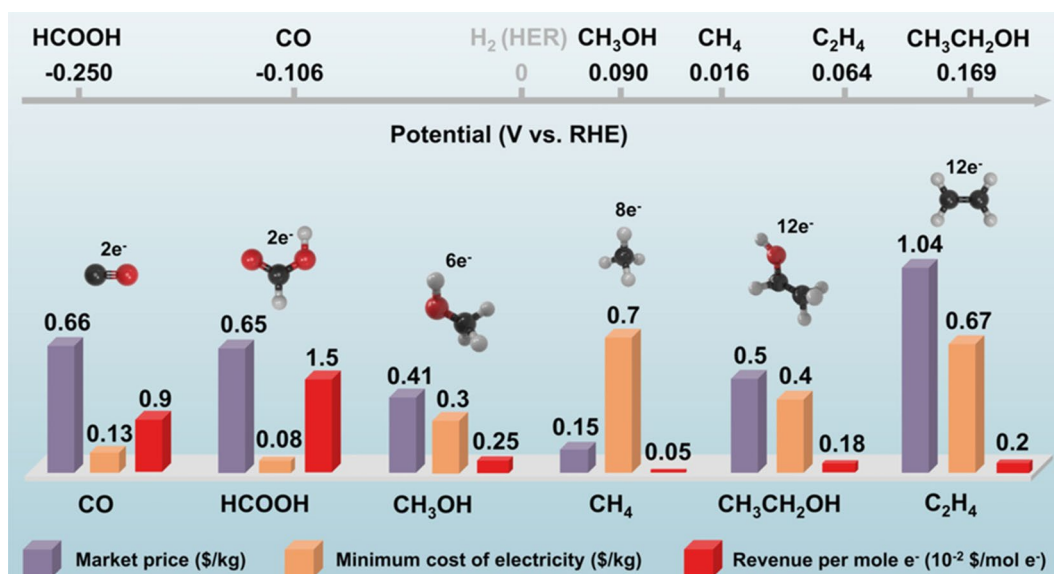
Moreover, CO<sub>2</sub> electrolysis can operate using renewables such as solar and wind as power source [84]. A solar photo-voltaic-driven electrolysis cell has been studied, resulting in solar to hydrocarbons (for example, CH<sub>4</sub>, C<sub>2</sub>H<sub>4</sub>) and oxygenates (for example, CH<sub>3</sub>CH<sub>2</sub>OH) efficiencies of 3.9 and 5.6% (with silicon solar cells and tandem cell, respectively), which turned out to be higher than that of natural photosynthesis [91]. Meanwhile, the solar to CO efficiency has increased from ~ 6% to ~ 19% over the past few years [92–94].

Table 1 shows selected reactions and reported Faradaic efficiency of the CO<sub>2</sub>RR leading to some of the C1–C3 chemical products discussed here.

Figure 9 shows the standard potentials of CO<sub>2</sub> reduction reactions and economic analysis of the products obtained

**TABLE 1:** Half reactions and Faradaic efficiency of the CO<sub>2</sub>RR resulting in C1–C3 products.

Product	Reaction	Faradaic efficiency (%)	References
Carbon monoxide (CO)	$\text{CO}_2 + 2\text{H}^+ + 2\text{e}^- \rightarrow \text{CO} + \text{H}_2\text{O}$	> 95	[3]
Methane (CH <sub>4</sub> )	$\text{CO}_2 + 8\text{H}^+ + 8\text{e}^- \rightarrow \text{CH}_4 + 2\text{H}_2\text{O}$	80	[82]
Methanol (CH <sub>3</sub> OH)	$\text{CO}_2 + 6\text{H}^+ + 6\text{e}^- \rightarrow \text{CH}_3\text{OH} + \text{H}_2\text{O}$	88	[95]
Ethanol (C <sub>2</sub> H <sub>5</sub> OH)	$2\text{CO}_2 + 12\text{H}^+ + 12\text{e}^- \rightarrow \text{C}_2\text{H}_5\text{OH} + 3\text{H}_2\text{O}$	52	[87]
Formic acid (HCOOH)	$\text{CO}_2 + 2\text{H}^+ + 2\text{e}^- \rightarrow \text{HCOOH}$	97	[89]
Ethylene (C <sub>2</sub> H <sub>4</sub> )	$2\text{CO}_2 + 12\text{H}^+ + 12\text{e}^- \rightarrow \text{C}_2\text{H}_4 + 4\text{H}_2\text{O}$	80	[86]
<i>n</i> -Propanol (C <sub>3</sub> H <sub>7</sub> OH)	$3\text{CO}_2 + 18\text{H}^+ + 18\text{e}^- \rightarrow \text{C}_3\text{H}_7\text{OH} + 5\text{H}_2\text{O}$	30	[88]



**Figure 9:** Standard CO<sub>2</sub> reduction potentials and economic analysis of products from the reactions, taking into account market prices, minimum cost of electricity per kg, and the revenues per mole of electrons consumed. Reprinted with permission from [81]. Copyright (2021) Wiley.

[81]. The analysis considers market prices, minimum cost of electricity per kg, and the revenues per mole of electrons consumed. The minimum cost of conversion into CO and HCOOH is lower (0.13 \$ kg<sup>-1</sup> and 0.08 \$ kg<sup>-1</sup>, respectively) compared to the other products. Moreover, their revenue per mole of electrons consumed is higher, which make these chemicals suitable target products. However, considering the separation costs of liquid formate from the electrolyte, it has been concluded that the electrochemical reduction of CO<sub>2</sub> to CO has significant economic and technological advantages over other chemicals [81].

## Outlook

Despite the advantages of electrochemical energy conversion and storage systems over their fossil fuels counterparts (low-operating temperatures, higher efficiencies, zero or low emissions of GHG to the atmosphere, in most cases all-solid configuration without moving parts) there are several issues to be

overcome before their extensive commercialization. It should be mentioned that some electrochemical technologies have reached niche markets, but improvements must be made. Granting that each device has its own particular challenges, higher costs than conventional systems are a shared issue.

It is generally proposed that, to compete with fossil fuels systems, environmental and sustainable aspects must be considered. Under this view, the need to implement these green technologies is justified. Great efforts are being made internationally to achieve large-scale production, thus decreasing costs. Several countries have started large collaborative projects, investing significant amounts in public and private funding.

For instance, the hydrogen council mentions roadmaps already developed in 30 countries, with a projected total investment of \$300 billion through 2030 [96]. Companies are planning sixfold and 16-fold increases in their investments through 2025 and 2030, respectively [96]. Similarly, the European Commission, via the European Battery Innovation project has committed €2.9 billion funding in the next few years, which will be

supported by €9 billion in private investments, to innovate the entire battery value chain [97].

Therefore, it is expected that green electrochemical energy conversion and storage systems will play a more important role in the energy scenario, aiming to achieve a sustainable future.

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## Data availability

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## Code availability

Not applicable.

## Declarations

**Conflict of interest** The authors declare no conflicts of interest.

## Supplementary Information

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