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Perspective

Green Hydrogen Production by Anion Exchange Membrane Water Electrolysis: Status and Future Perspectives

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Abstract: Green hydrogen production, i.e., produced on a CO_2-neutral basis through the electrolysis of water employing renewable electricity, has attracted increasing attention. The electricity required is generated from Renewable Energy Sources (RES), for example, wind energy, hydropower, or solar energy. Since neither the process of production nor the end products of H_2 and O_2 are harmful to the environment, green hydrogen is climate neutral. Developing electrolysis technology is, therefore, a research topic to follow. Anion Exchange Membrane (AEM) Water Electrolysis (WE) is an innovative technology that couples the advantages of the more mature technologies of Proton Exchange Membrane (PEM) and conventional alkaline electrolysis, with the potential to eliminate the drawbacks of both. AEMWE technology is in an evolutionary stage and involves more investigation on several research topics, such as membrane and catalyst development and stability, as well as alternative feeding solutions that do not compromise the availability of fresh water. These topics are addressed in this paper, mentioning the state-of-the-art materials, new promising ones, and providing future research directions to improve AEMWE towards a most mature technology.

Keywords: green hydrogen; water electrolysis; anion exchange membrane; transition metal catalysts; durability; feeding solution

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1. Introduction

Considering the world's growing interest in producing green hydrogen, WE using RES appears as a promising solution. The two leading WE technologies to produce hydrogen are conventional alkaline and PEM electrolysis, where the ions crossing the diaphragm/membrane are hydroxide anions and hydrogen protons, respectively. The latter presents two main advantages: great flexibility (On/Off cycling and transient operation) and operation at high current densities. However, the high production cost related to the materials used (Platinum-Group Metals—PGM and titanium) is a limitation. Conventional alkaline WE with liquid electrolytes are notable among others due to their larger maturity and higher commercial outreach [1]. In the conventional configuration, a thick diaphragm is necessary to separate the electrodes with a liquid electrolyte, resulting in moderate current densities, lower hydrogen purity, and weak load cycling. Nevertheless, this technology allows the use of non-noble metals, reducing production costs, and has a higher lifetime. Recently, polymeric AEMs have been developed for electrochemical system applications [2]. They included advantages from both PEM and alkaline WE. The principal modification between conventional alkaline and AEMWE is the substitution of the diaphragm by an AEM, enabling the operation at higher current densities and preserving the opportunity of using non-noble metal catalysts. There is a research gap concerning the main components and integrated AEMWE that can be assigned to the system complexity and the lack of standard evaluation criteria. For instance, most of the Hydrogen Evolution Reaction (HER) and Oxygen Evolution Reaction (OER) electrocatalysts described with excellent activity are tested at room temperature, for a small period, at low voltages, but these conditions do not correspond to real applications [3]. To be a competitive technology, by 2050, AEMWEs must operate at current densities above 2 Acm^{-2} with a voltage below 2 V [4]. Accelerated Stress Tests (ASTs) are decisive for determining the targeted lifetime of AEMWEs. Developments regarding the key electrolyzer components, such as the membrane, electrocatalyst, and their effects on electrolysis performance, have been reported in the literature [5]. The Membrane Electrode Assembly (MEA) is the main component that determines the performance of the electrolyzer, comprising the AEM, ionomer, anode, cathode catalyst layers, and Gas Diffusion Layers (GDLs). Although some membranes are commercially offered, they have two important weaknesses: the mobility of anions through the AEM is considerably lower than the mobility of protons through a PEM, and they do not demonstrate good chemical and/or mechanical stability. The alkaline media brings the advantage of using a wider variety of cheaper materials for AEMWE components compared to PEMWE. The feeding-water quality plays a vital role in the electrocatalytic activity and significantly affects the performance of the AEMWE. However, the use of impure water brings a great research opportunity considering the shortage of drinking water, but technical barriers for impure-water splitting need to be overcome [6].

There are several reviews available in the literature concerning AEMWE technology from a global perspective [2–4,7–10] or addressing specific topics such as membranes [5,11,12] or electrocatalysts [13], which could be useful to readers who desire to deepen their knowledge of the topic.

In this perspective paper, four research lines considered hot topics were selected: membrane and catalyst development, and durability and feeding solutions options, including the interesting possibility of using wastewater.

2. Hot Topics

2.1. Membrane Development

The AEM, comprising an anion exchange cationic group linked to a polymer backbone, is a key constituent that determines the AEMWEs' performance and lifetime. The mechanical and thermal stability is related to the polymer backbone chemistry, whereas the ionic conductivity is influenced by the cationic functional groups. The chemical stability is influenced by both the polymer backbone and the functional groups [7]. There are several AEMs commercially available, and others developed "in-house", reported in the literature. The polymer backbone is frequently a polysulfone, crosslinked polystyrene with divinyl benzene (DVB), or polyaromatics [7]. For example, poly(fluorenyl aryl piperidinium)-, polycarbazole-, and polyphenylene-based AEMs were tested in cells attaining current densities as 2.3, 2.57, and 5.3 Acm^{-2}, respectively, measured at 1.8 V, presenting good durability [4]. The cationic functional groups are generally quaternary ammonium (QA) or imidazolium (IM). Some suppliers commercialize AEMs, such as: Tokuyama (A201) (discontinued production), Fumatech (FumasepR), Ionomer (Aemion^{TM}), Dioxide Materials (Sustainion®), and Orion Polymer (Orion^{TM}). There are research-based membranes, e.g., Versogen (PiperION) and Xergy (XION™ Ion Exchange Membranes), which are promising alternatives to overtake commercial membranes. Nevertheless, they are offered on the market, although they are costly due to their small production scale and are not cost-competitive with commercial ones. In addition, more studies regarding durability under severe conditions are needed [4].

Liu et al. [14] tested the performance of an AEMWE at 60 °C with different commercial membranes: Sustainion® 37-50, Fumasep FAS-50, Fumasep FAPQ, Neosepta ACM, AMI 7001, Nafion® 115, and Celazole® PBI. Better results were found using Sustainion® 37-50 membranes, operating at 1 Acm^{-2} at about 1.9 V for 2000 h. The other membranes presented a high area-specific resistance (ASR) or a higher degradation rate under the tested conditions.

Henkensmeier et al. [11] published a very interesting paper, giving an overview of the commercial AEMs (Fumatech Fumasep® FAA3, Tokuyama A201, Ionomer Aemion™, Dioxide Materials Sustainion®, and membranes commercialized by Orion Polymer, Cohoes, NY, USA), and reviewed their properties and performances in AEMWE. The authors concluded that all materials (membrane, ionomer, catalyst, cell design, etc.) must be optimized together. Consequently, it is difficult to precisely rank the commercial membranes by analyzing the available literature data, limiting an experimental rank to a system. As an overall tendency, however, the membranes FAA3 and A201 presented a lower ion conductivity than the novel membranes Aemion and Sustainion®.

Fortin et al. [15] tested Aemion™ AEMs (all-hydrocarbon) with several ion-exchange capacities and thicknesses. The authors reported that better results were obtained using the AEM AF1-HNN8-50, with a thickness of 50 µm, achieving current densities of 2 Acm^{-2} at a potential of 1.82 V using 1 M KOH at 60 °C.

Kang et al. [16] used an AEM without an aryl-ether backbone structure (Orion-TM1™). The authors reported that this membrane contains quaternary ammonium as a functional group and polyphenylene as the backbone, presenting a high ionic conductivity and stability, performing better and being more durable than the conventional AEM (FAA-3). Additionally, it showed high efficiency using pure water as a feed solution. The optimized TM1-MEA attained a current density of 2.75 Acm^{-2} at 1.9 V with great stability of 55 $mV\ h^{-1}$, benefiting from the superior structural advantages of the Orion-TM1™ membrane. The aryl-ether-free structure and hydrophobic properties inhibited decomposition by ether hydrolysis and prevented the attack of hydrophilic hydroxide ions.

Lv et al. [9] developed a quaternary ammonium poly (n-methyl-piperidine-co-p-terphenyl) (QAPPT) and Ni-Fe layered double hydroxide (LDH) composite membrane. The QAPPT/3% Ni-Fe LDH composite membrane exhibited a great performance of 1 Acm^{-2} at the voltage of 1.824 V at 60 °C, superior to most of the ones reported in the literature. Moreover, a long-term stability test was conducted at 500 mAcm−2 for more than 87 h, achieving a reasonable performance.

As explained in the introduction, the MEA, comprising not only the membrane but also ionomer, anode, and cathode catalyst layers and GDLs, needs to be optimized. A recent review [9] explained the most common methods used to produce MEAs, catalyst-coated membrane (CCM), and catalyst-coated substrate (CCS) methods. The commonly implemented hot-press method used in PEM cells was not a good option in AEMWEs because i) the degree of mechanical force and high temperature induces irreversible damage to the membrane due to the use of metallic GDLs, and ii) hydrocarbon ionomers do not have an accessible glass transition (cannot be effectively fused by hot-pressing). This may lead to performance losses and degradation generated by insufficient contact and/or gradual delamination between the membrane/catalyst layer interface. Catalyst–ionomer interaction and loading content optimization are crucial for the advances in AEMWEs [17]. Engineered MEAs must be developed, aiming for effective contact between membrane/catalyst/ionomer/GDL.

2.2. Non-PGM Catalyst Development

Theoretically, due to a high pH environment, AEMWE can permit the substitution of PGM electrocatalysts with Earth-abundant first-row transition metals (e.g., Mn, Fe, Ni, Cu, and Mo) to catalyze OER and HER reactions. Currently, IrO_2, Ni, Ni-Fe alloys, graphene, $Pb_2Ru_2O_{6.5}$, and $Cu_{0.7}CO_{2.3}O_4$ are used as OER catalysts, while Pt black, CuCoOx, Ni-Mo, Ni/CeO_2-La_2O_3/C, Ni, and graphene could be used as HER catalysts [2]. Ir and Pt could be replaced by Fe, Ni, Mn, Cu, and Mo, considerably decreasing the prices of the global system. PGM-free electrocatalysts are already used to catalyze OER, with Ni-based electrocatalysts, which are currently state-of-the-art. However, for the HER, the state-of-the-art electrocatalysts are still based on precious metals [4].

Sulaiman et al. [13] wrote a complete review paper with an overview of the latest electrocatalysts for HER and OER. The authors reported that transition metal catalysts with very good electrocatalytic properties can play a role in improving AEMWE performance. For instance, the NiMo-NH_3/NH_2 for HER coupled with a Fe-NiMo-NH_3/NH_2 for OER reached 1 Acm^{-2} at 1.57 V. Li et al. [3] illustrated that OER kinetics can be influenced by a higher catalyst loading at the anode, and a lower utilization rate, particularly when the concentration of hydroxyl ions in the electrolyte has diminished, and active sites on the catalyst have been blocked by the inevitable high pressure of gas evolved in the MEA. The authors also stated that Ni-Mo alloyed materials, Co_3O_4 nanocrystals, Fe, and Ni-Fe alloys are good candidates to improve OER performance; however, stability through a period of one thousand hours needs to be confirmed. Sulaiman et al. [13] reported that bifunctional (the same for HER and OER reactions) catalysts should be investigated, making a simpler electrode system. For OER, many of the reported studies demonstrated high activity and efficiency of transition metal-based catalysts, predominantly NiFe-based, however, the majority still present high overpotentials beyond 200 mV, at 10 $mAcm^{-2}$ [13]. The high overpotentials are associated with the slow and difficult OER mechanism [3]. Considering HER electrocatalysts, the use of transition metals is still challenging because most of these catalysts are not as active as Pt/C. HER is also slower in alkaline media than in acid media [18]. Consequently, to decrease the overpotential and improve the activity of HER transition metal catalysts, there is a need to understand their reaction mechanism related to the adsorption of water molecules and bond breaking. One possible approach, reported by Li et al. [3], is the addition of heteroatoms, such as N-doping, possibly lowering the adsorption-free energy, facilitating HER. In addition, their stability and efficiency must be confirmed for real-world applications.

2.3. Durability

Most existing AEMWE components, especially membranes and catalysts, are not robust enough considering industrial targets due to chemical and mechanical degradation. This scenario is prone to change with intensive research and a few promising alternatives.

The causes of AEMWE performance losses include membrane degradation. It is well-known that the hydroxide anions attack the positively charged cations in the AEM, neutralizing them [4]. This leads to a loss of anion-exchange functional groups and, consequently, a decrease in $OH-$ conductivity generating higher ohmic resistances [19]. Additionally, continued thermal and chemical degradation of ionomers could reduce the connection to the catalysts, decreasing the reaction rate [13]. The ionomer ionic conductivity is better using a high number of ion-exchange groups, but this situation intensifies the water uptake originating the ionomer dissolution at higher temperatures. The catalyst activity may also become worse through catalyst dissolution, particle coalescence (catalyst agglomeration), Ostwald ripening, support corrosion, and adsorption of ionomer moieties [7].

Motealleh et al. [20] demonstrated a good performance, considering industrial parameters using Sustainion® membranes. Durability tests exhibited a stable performance of 1 $A.cm^{-2}$ at 1.85 V with a degradation rate below 1 mV/h over 10,000 h, leading to an estimated lifetime of about 20 years. Moreover, the authors reported that it is possible to increase the mechanical stability of a Sustainion® membrane through the addition of zirconia to the polymer matrix.

Henkensmeier et al. [11] determined that stability is intrinsically related to the system. The authors pointed out that a reliable prediction of membranes' lifetime is necessary. Since durability tests usually need up to 10,000 h, it is necessary to develop ASTs, as was performed for PEM Fuel Cells. Durability tests using ASTs to determine components' aging are crucial to attaining the industrial standard requirements. Overall, despite stability being recognized as a bottleneck, there is weak knowledge about the lifetime of optimized systems.

Bearing in mind catalyst stability, they must have a lifetime of more than 1000 h and keep their efficiency when operating at high current densities and under repeated on-off cycles from RES [13].

2.4. Feeding Solutions

In AEMWE, deionized water, ultrapure water, 1 wt.% K_2CO_3, and 1 M KOH have all been used as reactants [3]. Hydroxide solutions are used as an alternative to pure water to feed AEMWE to increase the ionic conductivity of the AEM because resistance losses decrease almost linearly (in a limited range of concentrations) when increasing electrolyte concentration [21]. However, higher hydroxide concentration conducts in elevated corrosion rates, reducing the lifetime of AEMWEs [7]. Better performances have been reported using 1 M KOH [2]. The predicted hydrogen demand in the distant renewable future is 2.3 Gt per year [22]. Considering the reaction stoichiometry, for every kg of hydrogen produced, 9 kg of water must be consumed. Therefore, 2.3 Gt of hydrogen requires 20.5 Gt, or 20.5 billion m^3 of water [23]. Although in other sectors, i.e., agriculture, more water is used than necessary for the most ambitious scenario for hydrogen, the scarcity of freshwater limits its use. As mentioned before, the feeding condition significantly impacts on the electrocatalytic activity and, consequently, AEMWE performance.

The use of demineralized water is the most preferred option to make AEM electrolysis a competitive technology. Nevertheless, using pure water carries critical new challenges. Ni is currently the standard material for electrode construction in AEM electrolysis. However, under anodic polarization, it is stable only at a pH higher than 9, conditioning the use of a pure water feed without performance degradation. Another problem with using deionized water, also occurring using hydroxide solutions, is the negative influence of CO_2 on performance. Parrondo et al. [24] described that this phenomenon was due to dissolved carbonate and bicarbonate anions that diminish the ionic conductivity of the membrane and catalytic layer binders.

The use of impure water in AEMWE is also an important research topic. Even if most pollutants in wastewater are successfully removed by the current wastewater treatment processes, small quantities of impurities (residual organics and ions) remain in the treated water. No studies show how the impurities affect water electrolyzer performance and durability, despite studies reporting how impurities affect either the membrane or electrode. There is a need to understand more fully the impact of water impurities present in wastewater on the performance and durability of AEMWE. For example, despite the fact that most cationic impurities have undesirable effects during electrolysis, cationic Fe impurities seem to be essential to stabilize OER activity in alkaline conditions with Earth-abundant catalysts [6]. Zou et al. [25] reported that Fe cations intensely improve OER activity when incorporated substitutionally into Ni or Co (oxy)hydroxides, acting as possible OER active sites. However, pure Fe (oxy)hydroxide is not a good catalyst for OER, and its effect still needs to be better understood. Cation poisoning can be avoided by adjusting the water flow rate and current density to minimize co-ion transport or through engineering approaches. It is also important to understand the relationships between the morphology, composition, and phase of the catalyst material and its durability in impure water AEM electrolysis. Analysis of Chemical Oxygen Demand (COD), Total Organic Carbon (TOC), and several metals (i.e., zinc, copper, iron) present in treated wastewater will be necessary to identify its composition. If CO_2 (previously identified as a detrimental compound) is present, it must be removed before using the wastewater. It is also possible that supplementary water treatments will be needed to eliminate specific impurities affecting AEMWE performance and durability. For example, precipitation or adsorption treatments could be able to remove undesirable metals.

Another possible solution is the use of seawater to feed AEMWEs. However, this latter approach leads to a side reaction, competitive with OER: the chloride oxidation reaction (ClER). Park et al. [26] thoroughly explained this competition and how to avoid/minimize it. The authors suggested that to achieve high OER selectivity, seawater electrolysis should be performed below 1.72 V_{RHE} under alkaline conditions, and a more active electrocatalyst for the OER is essential to attain highly efficient seawater electrolysis in an overvoltage range of less than 490 mV.

Few studies reported the use of other feeding solutions than hydroxide solutions.

Park et al. [26] tested an AEMWE using a Ni-doped FeOOH catalyst, tested in 1.0 M KOH + seawater, demonstrating a current density of 729 mA cm^{-2} and an energy conversion efficiency of 76.35%. Compared to reported AEM electrolyzers operated in 1.0 M KOH, the Ni-doped FeOOH operated in 1.0 M KOH + seawater still exhibited a higher performance.

Lindquist et al. [27] performed a study with an AEMWE made of commercially available materials, operating near 1.9 V at 1 A cm^{-2} in pure water. This study highlights that improving hydroxide conductivity, mass transport, and long-term ionomer stability are crucial for developing pure-water AEM electrolyzers.

3. Conclusions and Perspectives

AEMWE is a technology with tremendous potential for development, being the major challenge to overcome their high alkaline degradation. The main conclusions and perspectives are summarized below for each of the hot topics addressed.

Membranes:

Commercial membranes are available, differing in chemistry and thickness, that may contribute to system improvement and accelerate the research pathway. In particular, Sustainion® membranes constitute a promising option due to their good performance and durability. Different combinations using diverse anion exchange cationic groups linked to different polymer backbones must be investigated to develop more conductive, stable, and durable membranes. Moreover, all the AEMWE components should be optimized together, and experimental characterization recurring to IV curves, EIS, CV, and in situ/ex situ catalysts' characterization should be performed for a deeper understanding of components' interactions and the contribution of each component for performance losses and durability.

Catalysts:

Transition metal catalysts are already used with good results in OER, although few studies have been conducted reporting their use for HER. An interesting option leading to a simpler MEA construction is the use of bifunctional electrodes presenting good performances, considering both OER and HER reactions. Consequently, it is essential to further investigate the reaction mechanisms to a deep knowledge of the internal properties of these catalyst materials facilitating the development of more efficient catalysts (with lower overpotentials). Regarding the catalysts, deposition in the membrane (CCM approach) is preferable to guarantee better adhesion and to permit the use of lower catalyst loadings. Nevertheless, further optimization is essential considering ink composition and guaranteeing an appropriate electrode–membrane contact during assemblage.

Durability:

The biggest concern when developing new materials, catalysts, and membranes, besides their performance, is undoubtedly their durability. Some materials appear to be promising options, but once tested under real operating conditions, they demonstrated poor durability. ASTs protocols, mimetizing the dynamic RES operating conditions, must be widely implemented, allowing a quick durability assessment of the materials.

Feeding solutions:

The most used feeding solution is a hydroxide solution due to an improvement in AEMWE conductivity and, consequently, in performance. However, considering the shortage of fresh water, the desired options are seawater or treated wastewater. These options remain an ambitious scenario since AEMWE components quickly deteriorate with contaminants presented in impure waters, and/or the performance is lower due to competitive reactions occurring in the electrodes. More efforts in developing more robust and less susceptible to feed-water impurities AEMWE systems are mandatory to improve their lifetimes and reduce the overall cost.

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