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Research Trend in Anion Exchange Membrane Water Electrolysis System

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Abstract

In this paper, we have examined the key developmental issues and corresponding research trends of the anion exchange membrane electrolysis system. This system is regarded as the most economically viable and efficient among the current green hydrogen production methods, although it is still in the stage of research and development. Regarding the electrode catalyst, to capitalize on the advantages of utilizing non-precious metal materials under alkaline conditions, it is imperative to address both the enhancement of oxidation electrode catalyst performance and the design of electrodes with long-term stability. In terms of the ion exchange membrane used as the electrolyte, it should demonstrate low ion conductivity and improved durability in alkaline environment. Moreover, there is a growing demand for the development of innovative membrane-electrode assembly structure that can optimize the performance of the system components.

본 논문에서는 현재 그린수소 생산방식 중 가장 경제적이고 효율적인 시스템으로 여겨지나, 아직 기술 상용화 연구개발 단계에 있는 음이온 교환막 수전해 시스템의 주요 개발 이슈와 이에 따른 연구 동향에 대해 살펴보았다. 전극 촉매의 경우 알칼리 조건에서 비귀금속 재료를 적용할 수 있는 강점을 살리기 위해서는 산화극 촉매 성능 개선과 더불어 장기 안정성을 고려한 전극 설계가 필요하며, 전해질로 적용하는 음이온 교환막은 낮은 이온 전도성과 알칼리 환경에서의 내구성을 상승시켜야 한다. 또한, 구성요소들의 성능을 극대화할 수 있는 새로운 구조의 막-전극 접합체 개발이 요구된다.

Keywords

음이온 교환막(Anion Exchange Membrane),
수전해(Water Electrolysis),
전기화학촉매(Electrocatalyst),
막-전극 접합체(Membrane-Electrode Assembly)

1. Introduction

Hydrogen, although the most abundant element in the universe (making up 99 % by number of atoms and 75 % by mass), exists only in the form of compounds, such as water or hydrocarbons, due to its high reactivity and lightweight. Consequently, in order to produce hydrogen, it is necessary to separate it from these compounds, and during this process, other forms of energy (such as solar energy, heat, electricity, or nuclear power) need to be input to process (or convert) and produce pure hydrogen. Therefore, hydrogen is considered an energy carrier rather than a primary energy source, like fossil fuels that can be mined. Thus, along with the promotion of the fuel cell industry, which is the device that converts hydrogen into final energy, it is also important to consider how and to what extent hydrogen, which will be used in fuel cells in the future, is produced[1].

In addition, hydrogen offers various advantages as an energy supply source for military weapon systems, including naval ships and submarines. It can provide high-density energy required for various military missions, including not only conventional long-distance voyages but also advanced unmanned composite systems' energy sources. Additionally, hydrogen offers environmentally friendly and quiet operation, making it conducive to innovative application in future naval operations, thereby con-

tributing to military aspects through the activation of hydrogen technologies. With the aim of promote the activation of such hydrogen economy, it is required to make efforts in technological development across the entire cycle of the hydrogen economy, including production, storage, and utilization. Thus, the first and crucial step in this process is to ensure an adequate supply of hydrogen. While South Korea has achieved global competitiveness in the field of hydrogen utilization[2], such as hydrogen vehicles like the “NEXO,” there is inadequacy in the development of hydrogen production. Therefore, there is an urgent need for research and development in these areas.

Hydrogen can be classified into gray, blue, and green hydrogen depending on the feedstock and production methods. Among them, green hydrogen refers to environmentally friendly hydrogen that is generated by water splitting using renewable energy sources like solar power or wind power, without any greenhouse gas emissions. Although, the majority of current hydrogen produced is gray hydrogen, which has the lowest production cost. However, there is an increasing expectation for the production of green hydrogen to expand in order to achieve carbon neutrality. In other words, completely CO₂-free hydrogen is only possible through electrolysis, specifically green hydrogen production. This electrolysis technology is receiving significant attention as the most environmentally friendly technology, but securing its economic viability remains a challenge.

Electrolysis can be roughly categorized into three main types: alkaline water electrolysis (AWE), polymer electrolyte membrane water electrolysis (PEMWE), and anion exchange membrane water electrolysis (AEMWE) depending on the type of electrolyte used (Fig. 1)[3]. Among them, AEMWE combines the operating environment of AWE with the structure of ion-conductive PEMWE, aiming to overcome the shortcomings of existing electrolysis technologies. However, AEMWE is not yet the most mature technology and still faces numerous technological challenges.

Therefore, this mini review aims to examine the key issues and research status of AEMWE, which serves as a pivotal technology in the production of green hydrogen via electrolysis, and derive strategic implications for achieving carbon neutrality.

2. Electrode

2.1. Electrochemical/thermodynamic principles

AEMWE, a technology that combines the operating environment of AWE and the ion-conductive polymer electrolyte membrane structure of PEMWE, is considered as a next-generation technology that addresses the drawbacks of existing electrolysis technologies[4]. The electrolyzer, where water electrolysis (WE) takes place, comprises two electrodes, with the cathode facilitating the hydrogen evolution reaction (HER) and the anode facilitating the oxy-

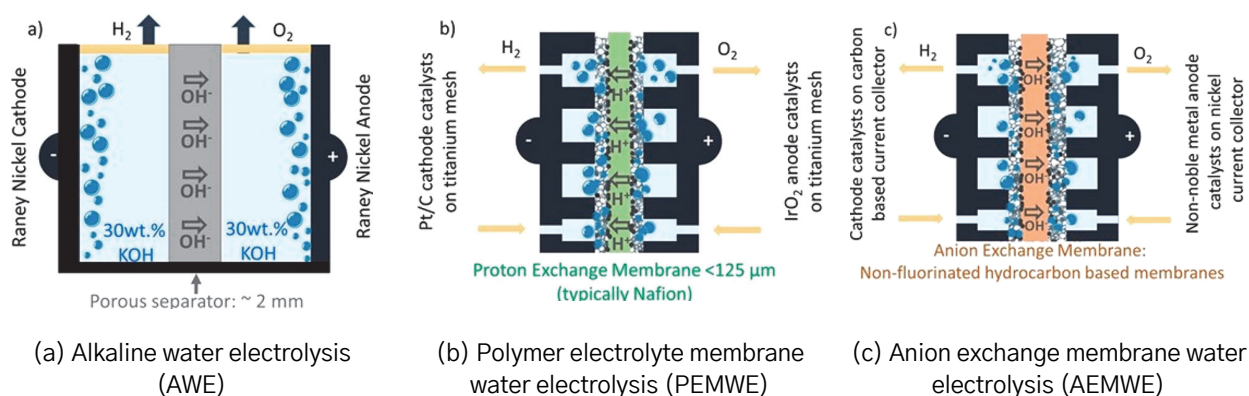
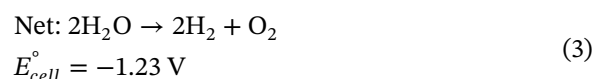
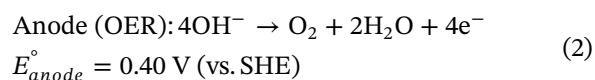
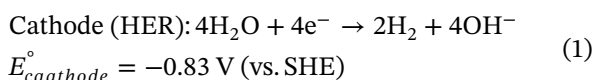


Fig. 1. The type of water electrolysis system (reprinted with permission from Open Access article [3])

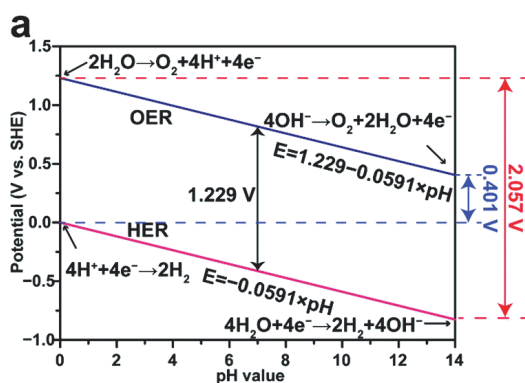
gen evolution reaction (OER), along with an anion exchange membrane (AEM) that allows the transfer of anions. It possesses a structure similar to that of PEMWE and fuel cells, facilitating the application of existing technologies. For WE, when the electrolyte is alkaline, OH^- ions serve as conducting ions, while in acidic electrolytes, H^+ ions fulfill this role. However, the decomposition reactions of water in both acidic and basic environments exhibit general similarities (Fig. 2). In the case of AEMWE or any electrolysis using an alkaline-media electrolyte, the reactions occurring in the electrolyzer are as follows:



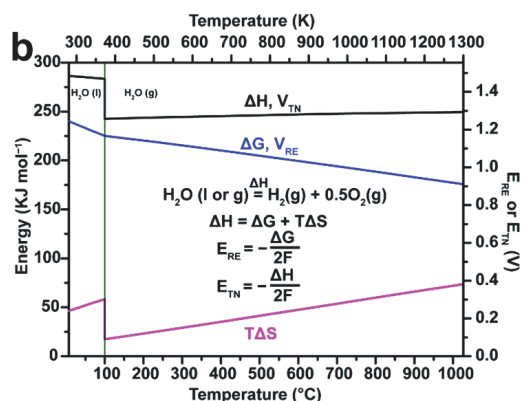
As evident from Eq. (3) above, the net voltage (E_{cell}°) is -1.23 V , and the negative sign signifies that the reaction is not a spontaneous reaction like a voltaic cell. In other words, the aforementioned reaction exhibits an endothermic nature, necessitating a cell voltage surpassing 1.23 V in order for it to

proceed. In the case of WE reaction, the standard enthalpy and entropy values are measured to be 285.84 kJ/molH_2 and 163.6 kJ/molH_2 , respectively. This notable contrast signifies a substantial alteration in the entropy of the reaction system as the liquid H_2O undergoes a transformation into the gaseous products H_2 and O_2 . Electrolysis at higher temperatures ($> 100 \text{ }^\circ\text{C}$), when water vapor is specifically employed as a reactant, the energy demands of the electrolysis process are diminished, as the elimination of these entropy variations are observed. Additionally, reference is made to the thermoneutral voltage (E_m°) in this context, which is 1.48 V for WE reaction. It represents transition point, known as the thermoneutral point, marks the boundary between endothermic and exothermic reactions, representing the potential at which the reaction advances without the need for external heat input. However, in actual electrolysis cells, E_m° is slightly higher than the theoretical value due to thermodynamic irreversibility and heat losses. To summarize, in low-temperature WE like AEMWE, additional voltage is required to consider overpotential and internal resistance losses for actual water splitting. The overall cell voltage can be expressed as follows.

$$E_{\text{overall}} = E_{\text{cell}}^\circ + \eta_a + \eta_c + iR_{\text{cell}} \quad (4)$$



(a) Thermodynamic potentials of the hydrogen evolution reaction (HER) and the oxygen evolution reaction (OER) in aqueous electrolytes with different pH values



(b) Thermodynamics of water splitting as a function of temperature.

Fig. 2. Thermodynamic parameters of water electrolysis according to electrolyte pH and temperature (reprinted with permission from Open Access article)[4].

In Eq. (4), η_a and η_c represent the overpotential at each electrode, indicating the difference between the equilibrium potential and the applied voltage required for the actual reaction. The term iR_{cell} signifies the ohmic drop.

2.2 Hydrogen evolution reaction (HER)

The specific reactions in hydrogen production can vary depending on the electrolyte type, but the generation of hydrogen through the adsorption and combination reactions of hydrogen ions on the electrode surface remains a similar mechanism. In alkaline media, kinetic mechanism underlying the HER encompasses the involvement of the Volmer step as the first process, where hydrogen atoms adsorb on the catalyst surface as intermediate species in the H^* state. Subsequently, H^* can either combine with other surface H^* (Tafel step) or react with H_2O (Heyrovsky step) to produce hydrogen. Therefore, the adsorption energy of hydrogen atoms on the catalyst surface is regarded as a crucial factor that significantly influences the performance of the HER. In alkaline media, as water is the sole proton-donating species, dissociation of H_2O in the Volmer step generally occurs slowly, making the initial water dissociation the rate-determining step [5].

Although the HER is widely recognized as one of the electrochemical reactions that has been extensively investigated and studied in scientific research, the alkaline HER involves relatively more complex reactions compared to acidic media. Therefore, HER catalysts that can stabilize OH^- and H^+ ions in water are highly advantageous, with bifunctional catalysts capable of stabilizing both species being particularly favorable. The HER activity exhibits a monotonous decrease as pH increases, supporting the theory of catalyst activity inhibition by higher-order hydrogen adsorption [6]. Additionally, HER occurs at more negative potentials in comparison to the OER. As a result, HER catalysts can utilize materials that are more stable compared to OER catalysts. The rela-

tively less demanding conditions of the HER allow for a wider range of options in terms of highly conductive and support materials with a large surface area, as opposed to the acidic-based PEMWE. In alkaline environments based on AEM electrolytes, inexpensive materials such as metal oxides, sulfides, and phosphides can be used as alternatives to precious group metal (PGM)-based catalysts, overcoming limitations of finding substitutes for PGM-based catalysts in acidic-based electrolysis [7]. For HER, extensive research has been conducted on PGM and/or non-PGM-based materials, which have demonstrated the highest intrinsic activities for the HER in electrolytes with a wide range of pH [8]. Furthermore, incorporation of nickel (Ni) into platinum (Pt)-based catalysts like alloys and Ni deposits on Pt surfaces, have exhibited the capability to surpass the HER activity of pure Pt under alkaline environment [9]. The synergistic interplay between Pt and Ni has been recognized to enhance HER. Ni, being an abundant metal and widely used as HER and OER catalyst in conventional water electrolyzers, has emerged as a fascinating contender to potentially replace catalysts based on Pt or Ru in alkaline environment. In order to enhance the catalytic activity, prevent hydride formation, and achieve enhanced stability, various strategies have been explored to stabilize the activity of pristine Ni catalysts. These include the formation of binary and ternary alloys of Ni with elements such as cobalt (Co), iron (Fe), molybdenum (Mo), cerium (Ce), zinc (Zn), and copper (Cu) [10].

2.3 Oxygen evolution reaction (OER)

For OER, direct generation of O_2 is challenging due to the high thermodynamic barrier in the combination process of intermediate species, O^* [11]. For instance, under alkaline conditions, to initiate the OER, hydroxyl radicals must first adsorb on the active sites of the catalyst surface to generate OH^* . Then, OH^* needs to undergo the removal of H^+ and

electrons to produce O^* , which subsequently reacts with OH^* to form OOH^* . Finally, through additional proton-coupled electron transfer processes, a single oxygen molecule and a vacant active site are generated [12]. In other words, as described earlier, the OER involves heterogeneous reactions between intermediate species containing oxygen, such as OH^* , O^* , and OOH^* , and four electrons, making it significantly sluggish compared to the HER, which involves the participation of only two electrons. Therefore, the OER is considered a bottleneck in the overall water splitting reaction. However, the detail reaction mechanism still remains uncertain, and further research is required to investigate the influence of electrolyte pH and operating conditions on the catalyst's activity and stability.

In particular, in the case of acidic conditions in PEMWEs, only PGM-based OER catalysts have demonstrated the necessary stabilities for practical applications. However, it is believed that the elevated pH environment in AEMWEs provides a wider range of possibilities for non-PGM OER catalysts, including transition metals (TM) such as Ni, Fe, Cu, and Co. Thus, electrochemical catalysts based on TMs such as Fe and Ni have been developed and commercialized for HER/OER due to these advantages. However, this perspective is oversimplified, as long term durability remains a challenge for many TM catalysts include their supports [13]. As a result, the available choices for materials suitable as OER catalysts and supports are constrained. Although IrO_2 and RuO_2 are regarded as cutting edge catalysts for the OER in acidic electrolytes, their long-term stability in high pH conditions is found to be inadequate [14].

Many improvements have been made to non-PGM OER catalysts in alkaline environment over the past decade. Among the most promising non-PGM catalysts for OER, Ni- and Fe-based catalysts are recognized for their high mass activity and low onset potential. Additionally, other alternatives such as Co and Cu have garnered interest for early studies

on AEMWE. Nevertheless, the evaluated mass activities and performance of AEMWE of these catalysts were lower than Ir-oxides or Ni-Fe-based catalysts. However, both Ni-Fe and Ni-Co-Fe-based catalysts have demonstrated comparable activities to Ir-oxides-based catalysts in both half-cell and full-cell configurations. Nonetheless, the stability of these Ni-Fe and Ni-Co-Fe-based catalysts still requires further improvement [15].

3. Anion exchange membrane

3.1 Issue: Low ionic conductivity and durability

In order to address the obstacles posed by the comparatively low ion conductivity and limited durability of AEMs in AEMWE, it is crucial to develop optimal membranes that demonstrate exceptional characteristics. These include high ion conductivity, superior thermal and mechanical stability in high-temperature and alkaline environment, along with low gas permeability. The AEM serves as a critical component of AEMWE and plays a crucial role in determining the operating environment of the cell, including temperature and humidification conditions. It facilitates the transfer of OH^- ions to the anode, which generated in the cathode. Interestingly, despite OH^- ions sharing the same ion transport mechanism (Vehicle or Grotthuss hopping mechanism) as H^+ ions, the ion conductivity of OH^- in AEMs is reported to be more than 56 % slower than that of H^+ ions ($198.3 \text{ S cm}^2 \text{ eq}^{-1}$ compared to $349.8 \text{ S cm}^2 \text{ eq}^{-1}$, respectively) [16].

Numerous research studies have focused on addressing the issue of low OH^- ion conductivity. These studies have explored methods like increasing the ion exchange capacity (IEC) of ion exchange resins or utilizing the microphase separation phenomenon to create permeation channels. Initially, early-stage research on AEMs aimed to enhance the ion conductivity of electrolytes by increasing the IEC to ensure a greater number of charge carriers. However,

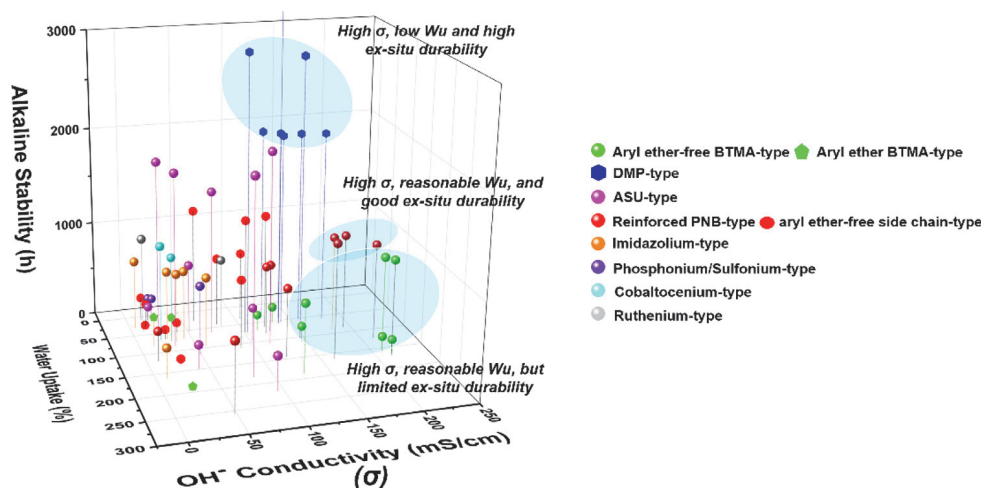


Fig. 3. Comparison between the water uptake (W_u), OH^- conductivity (σ), and ex-situ stability of various AEMs in current research (reprinted with permission from Open Access article [19]).

high IEC membranes inevitably lead to high water uptake, resulting in significant swelling and compromising the mechanical properties of the membranes (Fig. 3). This pronounced swelling behavior poses a significant drawback, as it raises the risk of membrane tearing or cracking during the assembly and operation of the humidification device in system. Moreover, the implementation of these electrolytes in the membrane-electrode assembly (MEA) can intensify the interfacial resistance between the membrane and the electrode, which in turn can result in electrode delamination [17].

3.2 Performance-enhancement strategies

In recent research, reinforcement composite membranes incorporating porous supports or block copolymer-based architecture capable of inducing microphase separation have been investigated as strategies to mitigate swelling and preserve mechanical properties. Moreover, attempts are underway to improve stability through the utilization of cross-linking techniques, which create a three-dimensional network resembling a mesh and establish connections between two atoms or polymer chains [18].

Reinforcement composite membranes are manufactured through two methods: impregnation of mechanically durable porous supports with ion-

exchange materials and infiltration of synthesized polymers, as well as pore filling with monomers followed by synthesis. In either approach, the swelling caused by ion exchange species is controlled by the outer walls of the porous structure, effectively maintaining the mechanical characteristics of the entire AEM. Block copolymers possessing microphase separation capabilities have demonstrated superior ion conductivity compared to polymers with equivalent IECs. This phenomenon can be attributed to the appropriate microphase separation between hydrophobic polymer backbone and the hydrophilic ion-conducting head groups, akin to Nafion, which facilitates anion separation within the hydrophobic domains. The development of hydrophilic domains for ion conduction allows for enhanced utilization of cationic regions, leading to high ion conductivity even at relatively low IEC values. Utilizing block copolymers in AEMs enables accelerated ion conduction through interconnected hydrophilic channels and water molecules, in contrast to the random distribution of ion exchange species in conventional ion exchange membranes [19]. Moreover, the segregated structure itself can be considered a form of microphase separated reinforcement composite membrane, where the hydrophobic segments function as a porous structure, effectively mitigating the mechanical degradation associated with high IEC.

To advance the field of AEM design, a systematic exploration of different microphase separated structures at the molecular level is necessary. This includes the use of predictive modeling and multi-scale molecular simulations to analyze species transport and water solvation within these materials. Peralkylammoniums are anticipated to continue being one of the most extensively researched and stable cation classes. However, future research efforts will likely concentrate on all-carbon backbones, structures free of aryl ethers, and polybenzimidazole (PBI) architectures. It is crucial to consider the processability and cost of the materials as well [18].

Therefore, it is crucial to develop AEMs that exhibit improved OH^- ion conductivity and alkaline stability, while maintaining mechanical integrity; and minimizing water swelling effects. Future research efforts should prioritize addressing these challenges to enable the large-scale deployment of AEMWE systems.

4. Membrane-electrode assembly (MEA)

The membrane-electrode assemblies (MEAs) play a pivotal role as a crucial component in the system, where the actual electrochemical reactions take place. Thus, it can be considered as the core element responsible for the functioning of these devices. In the construction of a MEA, there are two primary techniques for catalyst deposition: the catalyst-coated membrane (CCM) technique, where the catalyst is directly applied onto the membrane, and the catalyst-coated substrate (CCS) technique, where the catalyst is deposited on a substrate. For AEMWEs, the substrate options typically include gas diffusion layers (GDLs) or porous transport layers (PTLs), which can be coated with the catalyst using wet routes. In these wet routes, a stable ink or slurry is prepared by combining catalyst particles and ionomer as binder with a proper solvent. The resulting mixture is then applied to the GDL support through

spraying or painting. These techniques, initially developed for fuel cells, have been optimized for both PEMWEs and AEMWEs, more recently [20,21].

The research about overall design of the MEA has received little attention in comparison to catalyst or electrolyte membrane, but it has a significant influence on determining the internal electrode morphology and the formation of the triple phase boundary (TPB), where electrochemical reactions occur (Fig. 4). The characteristics of the fabrication methods used for MEA production or the properties of the catalyst slurries (or electrode materials) can significantly impact the TPB and electrode morphology. Therefore, to translate the outstanding performance of next-generation catalysts and electrolyte membranes reported from the laboratory scale into practical devices of the factory scale, there is an urgent need for research focused on MEA structure optimization and design improvements.

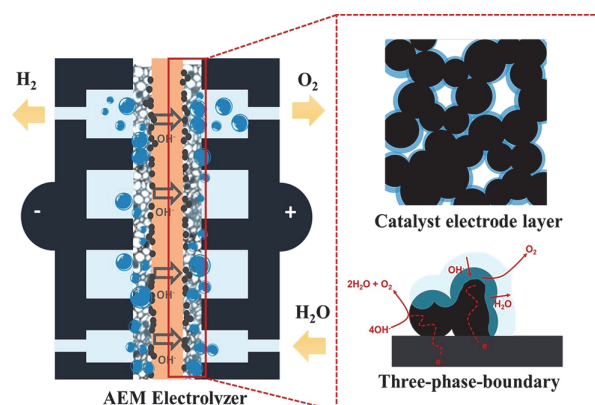


Fig. 4. Illustration of triple phase boundary for OER in MEA anode (reprinted with permission from Open Access article [3]).

Conventional methods used in the wet route fabrication of MEA have inherent limitations. These limitations include the production of a significant amount of waste in the form of catalyst ink slurries, which results in economic inefficiency. Additionally, these methods face challenges such as poor mass transfer, inadequate electrical conductivity, and performance degradation. In order to address these concerns, an alternative approach involves directly

forming catalysts on 3D-interconnected porous electrodes and omitting the ionomer [20]. This approach has the potential to enhance catalyst utilization and improve interactions between the catalyst and electrode (Fig. 5).

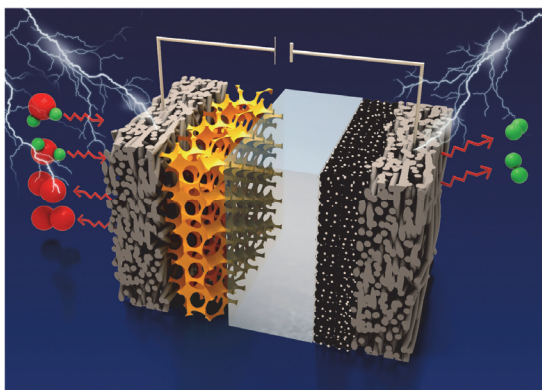


Fig. 5. Conceptual diagram of the ordered inverse-opal structured MEA (adapted with permission from [21], Copyright 2019 Elsevier.)

5. Conclusion

This comprehensive review focuses on analyzing the primary challenges and recent investigation tendencies of AEMWE systems, focusing on the different components, including electrodes, electrolytes, and MEAs. To achieve the widespread application of AEMWEs, it is essential to enhance the activity and stability of both the HER and the OER electrocatalysts. Urgent improvements in OER catalysts are particularly necessary due to the sluggish nature of the OER, which results in significant overpotentials. Extensive research efforts have been dedicated to acquisition a comprehensive prehension of the mechanisms underlying the OER, for develop more efficient catalysts with enhanced performance and stability. However, while gaining insights into reaction mechanisms is crucial, it is imperative to create catalysts that exhibit exceptional activity and stability within MEAs. Furthermore, the interaction between various membrane (ionomer) materials, such as quaternary ammonium groups and phenyl groups, predominantly used in membranes, and catalysts are a cru-

cial factor influencing the performance of AEMWEs. However, research on interfacial interactions is currently lacking.

Undoubtedly, although it is considered as a vague future technology, AEMWE possesses the potential for complete commercialization and can serve as a fundamental pillar for attaining green hydrogen. Moreover, AEMWE can serve as an efficient and effective energy source across the entire defense sector, encompassing naval applications as well. This can be accomplished through conducting focused research on the aforementioned issues and simultaneously developing innovative MEA structures that can deliver high performance in practical devices.

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