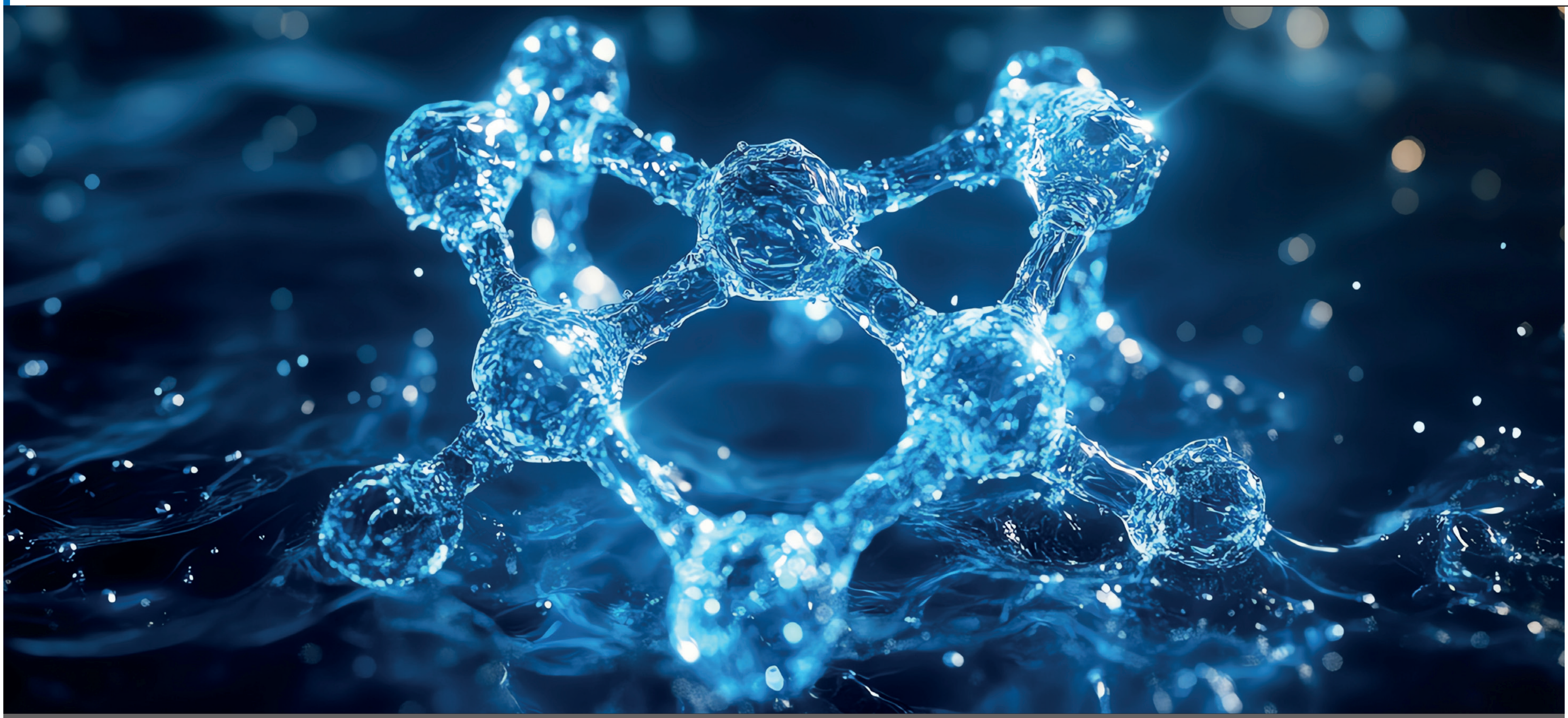

EXPLORING ALTERNATIVE APPROACHES TO HYDROGEN FUEL CELLS

Recent Advancements in Fuel Cell Technologies

Abstract

This article discusses recent research and advancement within the hydro fuel cell industry. This energy efficient and greenhouse-free renewable energy source holds significant potential for large scale deployment. However, limitations in key cell materials pose challenges to widespread adoption. This paper highlights advances in catalyst and membrane research. The industry is highly dependent on platinum-based catalysts, prompting efforts to reduce or replace platinum usage. The paper discusses the emergence of low-platinum catalysts, specifically Pt-Co, as well as non-platinum catalysts. Parallel studies catalysts being done investigating alternative membrane compositions, particularly non-fluoride options such as co-polynaphtoyleneimide and the hydrocarbon polymer p5PhSH. For both materials, the effects of sulfonation were studied to assess improvement in efficiency and proton conductivity, aiming to achieve performance comparable to fluoride membranes.

Introduction

Hydrogen fuel cells (HFCs) are a form of renewable energy that has been around for nearly 200 years, with the first prototype created in 1839 [1]. Since then, HFCs can produce significant amounts of energy that can power large-scale vehicles including trucks, buses and even trains [2]. A proton exchange membrane fuel cell (PEMFC) is how hydro fuel is produced. Figure 1 shows a conventional PEMFC [3]. The cell is composed of catalyst layers, gas diffusion layers (GDLs), and a membrane [4]. The catalysts layers are placed on opposite sides of the PEMFC. The catalyst for the anode layer is responsible for splitting hydrogen into protons and electrons while the catalyst for the cathode executes oxygen reduction reactions (ORR) to produce water. The GDLs promote efficient transportation of molecules in and out of the cell. The membrane is extremely critical to a PEMFC. It dictates what ions can pass through, separating the protons and electrons [4]. The emissions of PEMFCs are free of greenhouse gases, with their only byproduct being water. The eco-friendly nature and high energy conversion rates promote a promising future for HFCs [5]. However, materials that make up many of the PEMFCs today can be costly, limited, and have impacts on the environment [2]. Recently, research has been done on alternative catalysts along with different compositions of materials used in the membrane.

Catalysts

Catalysts play a vital role in hydrogen fuel cells. The separation of hydrogen into protons and electrons is lengthy and energy consuming, thus-necessitating fuel cells to require high amounts of energy for operation [6]. Noble metal-based catalysts are commonly used due to their efficiency and stability [6]. Platinum is one of the most widely used catalysts used in cells due to their high-performance capabilities. Platinum catalysts can be used for both the anode and the cathode components in the cell. The cathode requires much more platinum loading due to the substantial amount of energy needed for ORR. Platinum can handle the corrosive environment of a cathode while still maintaining a valuable catalyst performance [7]. Platinum is a useful and reliant component to hydro fuel cells. However, it is not the most attainable. Like many noble metals, platinum is scarce and can be expensive. With platinum catalysts responsible for 50% of the cell stack costs, scaling up in size to become commercialized can be quite challenging [8]. The recent advancements for catalysts involve lowering platinum usage or non-platinum alternatives.

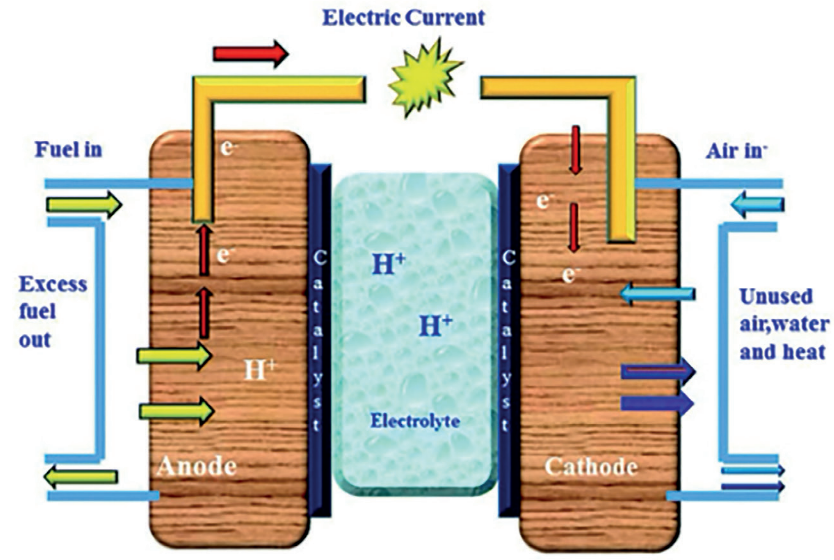

Figure 1: Diagram of PEMFC [3]

Low Platinum Usage

Platinum (Pt) can be alloyed with transition metals such as iron or copper which can lower Pt loading values. Utilizing transition metals can also facilitate oxygen dissociation and lead to higher efficiency [2]. However, a common result is harming the durability of the fuel cell from leaching of transition metals which causes corrosion of the cell. For platinum catalysts alloying with transition metals, a common pairing is with cobalt (Co) as Pt-Co. This is due to their ORRs that are higher than pure platinum catalysts. The activation of these catalysts also tends to be lower which can help lower costs [2,8]. In an experiment associated with University of California Irvine and by Chen et al., Pt-Co catalysts were experimented to see how the pairing can be optimized [2]. To do this, the catalysts were tested on two different membrane electrode assemblies (MEAs). Shown in Figure 2, one was with Vulcan carbon and the other a high surface area carbon (HSC). Both systems were put through accelerated stress tests (ASTs). which were used to simulate conditions as if they were used in large vehicles. Specifically, it increased temperatures to around 90° C as well as pressure to about 250 kPa. For degradation of MEAs, both had electrochemically active surface area losses less than 40%. It was found that the high surface carbon MEA had the highest amount of electrochemically active surface area. However, the HSC was also the MEA to face the highest percentage of electrochemical surface area degradation. As Chen et al. pointed out, this could be due to the increase in crystal size of nanoparticles. As crystal size increases, the electrochemical surface area (ECSA) will also increase, leading to higher amounts of the catalyst facing oxidation and platinum reduction. For Volcan carbon, crystal size increased to 24% while HSC had an increase of 32%. Correlation between crystal size and loss of ESCA can be an explanation for this result. The study also focused on effects of Co leaching, which can decrease a catalysts electrochemical activity, referred to as mass activity (A mg-1). For Volcan carbon an 80% decrease in mass activity was found, for HSC the decrease was 66%. Degradation was expected as it is associated with alloying with transition metals [2]. Transitional metal platinum alloys are a promising area for fuel cells. It is paving ways to become less reliant on platinum while maintaining performance. Although this area is still critical for research, the jump to non-platinum catalysts has already been made [2].

Non-Platinum

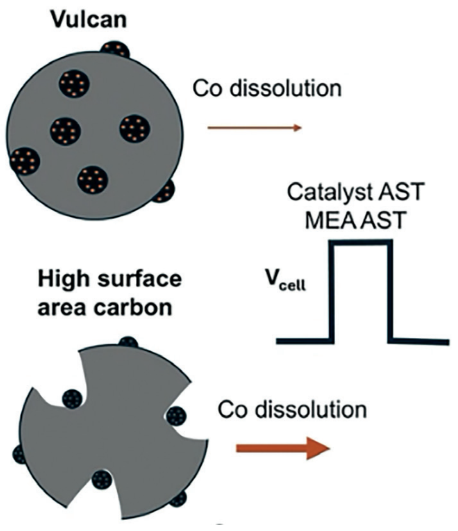

Figure 2: Diagram of MEAs used [2]

Metal-Nitrogen-Carbon (M-N-C) catalysts have potential to be industrialized and replace platinum catalysts used in cathodes [9]. There has been focus on methods to incorporate iron into carbon-based materials [10]. Figure 3 shown below is a representation of methods used to combine iron and carbon [10]. One method is through mechanical mixing which are classified as simplistic and inexpensive. This method typically produces non-uniform distribution of iron through carbon material. The self-assembly strategy controls chemical reactions to between iron and carbon that creates metal-organic frameworks (MOF) which can then be converted into catalysts. Iron-based catalysts through this method have shown improved uniformity of iron distribution as well as carbon size, overall enhancing the catalyst's productivity [10]. These differing techniques can be beneficial in improving the catalyst's performance.

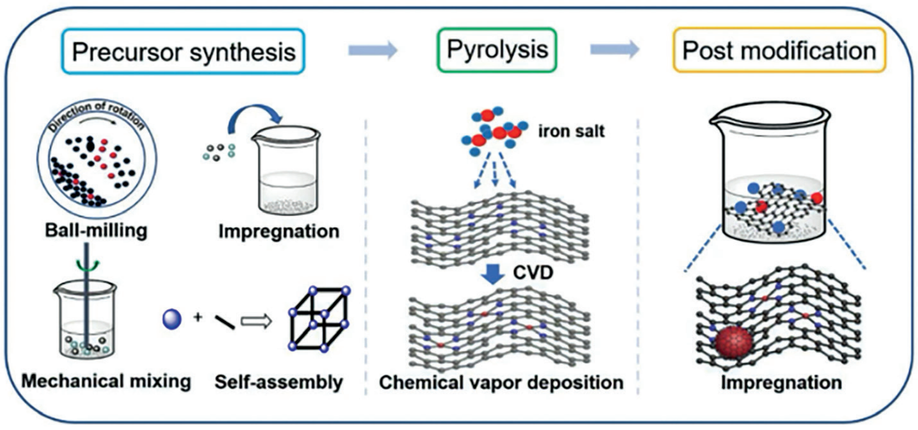

Figure 3: Methods for combining Iron and Carbon. [10]

Iron-nitrogen-carbon (Fe-N-C) catalysts appear to have high activity like that of a Pt-C catalyst [9]. However, stability is a major setback. In a study done associated with Argonne National Laboratory, Liu et al. made a focus on strengthening catalysts through a specific treatment process [11]. The process was primarily composed of two steps: a thermal treatment followed by a vapor deposition. For thermal treatment, NH4Cl and ammonium chloride were used on the iron catalysts which were subjected to temperatures ranging from 300-1100°C, the catalyst being referred to as Fe-AC. It was observed that through this treatment ORR can be achieved between 400°C and 500°C but is optimal at 700°C. For the second part of the treatment, chemical vapor deposition (CVD) was used. For this to be executed, zinc-based zeolite imidazole framework (ZIF-8) nanocrystal powder was applied as a thin layer on the Iron-Ammonium chloride catalyst, the produced catalyst was referred to as Fe-AC-CVD [11]. To see if the two-step treatment would be critical, the Fe-AC and Fe-AC-CVD catalysts were both used in MEAs. The current density signifies the rate of electrochemical reactions on a catalyst. Higher densities facilitate kinetic reactions and ultimately improve ORR efficiency. Both catalysts performed well with current densities of 44.2 mA cm−2 at 0.9 ViR-free and 33 mA cm−2 at 0.9 ViR-free, for Fe-AC and Fe-AC-CVD respectively. However, when each catalyst performed ASTs, there were noticeable differences. The catalysts without the CVD experienced a 94% decrease in density after 30,000 AST cycles. For the catalyst with the CVD applied, only 5.1% of its density was lost, ultimately having a similar performance of a Pt-C catalyst. Table 1 shown below, lists different catalysts along with some parameters [2,11,12]. The higher half-potentials correlate with favorable reactions that require lower activation energy [13]. Half-potentials also depend on catalyst loading rates, which is why evaluating other measurements such as current density is valuable.

Fluoride-Free Membranes

Polyfluoroalkyl and perfluoro sulfonic acid membranes are widely used in hydro fuel cells. PFSAs play a key role in the hydro fuel industry today. Their stability and proton conductivity adhere to the industry's dependency on them. Like platinum catalysts, one of the major drawbacks to PFSA membranes is the cost which is heavily influenced by the fluorine composition [14]. PFSA is composed of hydrofluorocarbon chain with extremely strong bonds that are responsible for some of the membrane's beneficial attributes [15]. However, the material raises concern for both human health and the environment [16]. While durability can be useful in producing fuel cells, the perfluorinated compounds released from the cells can linger in the environment. The compounds can end up in water sources and go undetected in water treatment plants, imposing great risk for human consumption. Reproductive issues, hormonal disturbances, and liver problems are some health concerns associated with perfluoro substances [16]. Researchers have focused on ways to eliminate its usage without sacrificing results.

Table 1: Catalysts and associated parameters [2,11,12]

Catalyst	Half Wave Potential (V)	Current Density ($A\ cm^{-2}$)	Degradation	Source
Pt-Co	0.87	0.8	14%, ~ 100 hours	[2]
Fe-AC	0.915	0.0442	94% loss, ~ 300 hours	[11]
Fe-AC-CVD	0.846	0.033	5.1%, ~ 300 hours	[11]
Fe-NC	0.838	0.147	--	[12]
Fe-NCBrCl	0.838	0.781	--	[12]

In a work supported by the Ministry of Science and Higher Education of the Russian Federation, Zavorotnaya et al. performed a comparative experiment between a widely used fluoride membrane, Nafion 212, as well as the non-fluoride membrane sulfonated co-polynaphtoyleneimide (co-PNIS), shown in Figure 4 [17]. The membranes were used to create MEAs that were then compared. For the operating power, both membranes carried out similar values, with 415 mW/cm2 and 419 mW/cm2 for co-PNIS and Nafion 212 respectively. There was roughly a 20% difference in values between the membranes for proton conductivity and maximum power, with Nafion 212 obtaining higher values. The results were met through optimization of co-PNIS' parameters. This includes temperature values which were found at 60-65° C, as well as pressure that was found at 13 psi. While these results do have some variations in values, a 20% difference between the fluoride and non-fluoride MEAs offers potential for co-PNIS capabilities [17].

At University of Pennsylvania, research on fluorine-free membranes has been funded by the Department of Energy. In a recent paper by Oh et al., sulfonated polymer membranes were tested on their performance such as stability and conductivity [18]. The experimented hydrocarbon polymer, referred to as p5PhSH, has phenylsulfonic acid groups attached to a polyethylene backbone. Performance of p5PHSH was tested through an all-atom molecular dynamic (MD) simulation, a computer analysis method for observing properties of molecules and atoms. It was observed through the simulation that p5PHSH performs good nanophases separation behavior; a critical factor that creates distinct hydrophilic and hydrophobic separation for efficient proton conductivity. Copolymers were created all ranging in sulfonate percentages which were followed by an all-atom MD analysis for water intake and efficiency. All the copolymers performed well with ion-exchange efficiencies (IEC) of 2-3.9 millimoles of sulfonate ions per gram of dry polymer (mmol/g) which increased as sulfonate percentages increased. The values exceed a commercialized membrane, Nafion NR211 that has an IEC of roughly 0.91 mmol/g. While the sulfonate percentage influences the IEC values, for proton conductivity a different conclusion was met. All sulfonate levels above 34% performed in a similar manner as shown in Figure 5 [18]. The influential factor in this parameter appears to only be hydration levels, to which increasing the water uptake by increasing relative humidity. The experiment concludes that sulfonate levels for p5PhSH can be optimized by analyzing nanoscale characteristics. Table 2 lists various membranes with corresponding values. Ultimately, p5PhSH has the potential to make an impact on the hydro fuel cell industry by providing a fluorine-free membrane alternative [3,18,19].

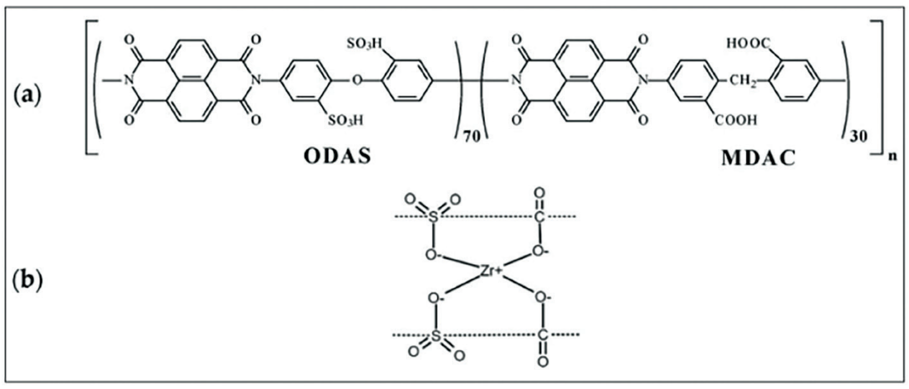

Figure 4: Chemical structures of (a) co-PNIS, and (b) Zirconium used for polymerization [17]

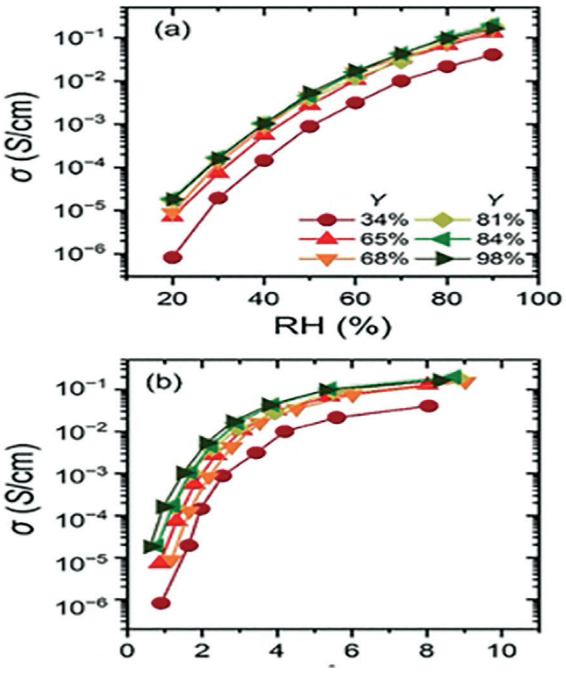

Figure 5: Proton Conductivity of p5PhSH [18]

Membrane	IEC (mmol/g)	Proton Conductivity	Source
NR211	0.91	0.180	[19]
SPEEK/PVDF/BP-10	1.38	0.039 (80)	[3]
SPEEK/SPVdF-HFP/S-SiO2 (6 wt%)	1.71	0.079 (90 °C)	[3]
p5PhSH-34	2.0	0.10 (50)	[18]
p5PhSH-68	3.4	0.10 (40)	[18]
p5PhSH-84	3.9	0.10 (40)	[18]

Conclusion

PEMFCs have shown great improvements in recent years. The studies for alternate catalysts, whether low-platinum or platinum-free, offer solutions to alleviate platinum usage in industry [5,11,12]. The M-N-C catalysts are gaining traction, specifically iron based catalysts. From the studies mentioned, it is apparent that non-platinum catalysts have the capabilities to perform as well as platinum-based catalysts. The two-step treatment by Liu et al. demonstrated the influence treatment processes have on a catalyst's performance. Fe-AC-CVD having capabilities to perform as well as platinum-based is a clear representation of that [11]. Catalysts are not the only component in PEMFCs that are seeking alternatives. Research into the composition of membranes, specifically the non-fluoride type, has approaching values in proton conductivity like industry used membranes [3,18]. The advancements of hydro fuel cells are necessary to grow the industry while making them affordable and sustainable.

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