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Nafion
Nanofillers
Composite
PEMFCs

Proton exchange membrane fuel cells (PEMFCs) and proton exchange membrane water electrolyzers (PEMWEs) are important technologies in hydrogen energy system. Perfluorosulfonic acid ionomers, particularly Nafion, serve as the benchmark solid polymer electrolyte due to their high proton conductivity and structural stability under fully hydrated conditions. However, Nafion experiences severe performance loss under elevated temperatures ($> 80^{\circ}\text{C}$) and low relative humidity because of membrane dehydration. Nafion is also susceptible to fuel crossover and radical induced chemical degradation. Incorporating nanofillers into the Nafion matrix offers a versatile strategy to overcome these operational limitations. This review discusses six principal classes of nanofillers: metal oxides, solid acids, carbon based nanomaterials, clays and zeolites, metal organic and covalent organic frameworks (MOFs/COFs) and polymeric/biobased additives. Their reported roles in hydration control, proton transport, fuel crossover suppression, mechanical reinforcement and chemical stability are critically compared.

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Global decarbonisation efforts are expected to increase hydrogen demand substantially, with projected to reach 560 million tons by 2050[1]. The growing deployment of Proton exchange membrane fuel cells (PEMFCs) contributes to hydrogen demand because these systems consume hydrogen to generate electricity, while Proton exchange membrane water electrolyzers (PEMWEs) supply hydrogen through electrically driven water electrolysis. These PEMFCs and PEMWEs represent two complementary applications of Nafion in hydrogen energy system as illustrated in Figure 1. Nafion is widely used as a solid polymer electrolyte in proton exchange membrane fuel cells (PEMFCs) and proton exchange membrane water electrolyzers (PEMWEs).

In a PEMFCs, the membrane conducts protons from anode to cathode while separating the reactant gases and preventing electronic conduction. Nafion transports the protons to the cathode, while the electrons pass through an external circuit to generate electricity. At the cathode, oxygen combines with these protons and electrons to form water. The membrane also separates the reactant compartments and restricts gas crossover. In PEMWEs, an external power supply drives water

oxidation at the anode, and the resulting protons migrate through Nafion to the cathode, where they are reduced to hydrogen. In both applications, the membrane must provide high proton conductivity while maintaining low gas permeability and adequate chemical and mechanical stability.

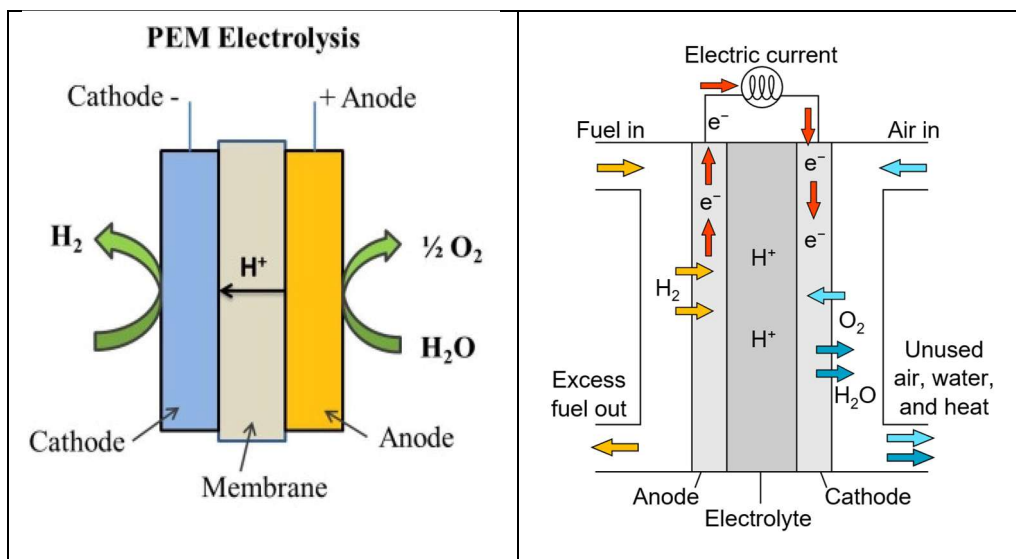


Figure 1: Schematic diagram of Proton Exchange Membrane Fuel Cell (PEMFC) and Proton Exchange Membrane Electrolyzer (PEME)

Nafion, a perfluorosulfonic acid ionomer with chemical structure as shown in Figure 2, has long served as a benchmark because of its high proton conductivity and chemical and mechanical stability [2].

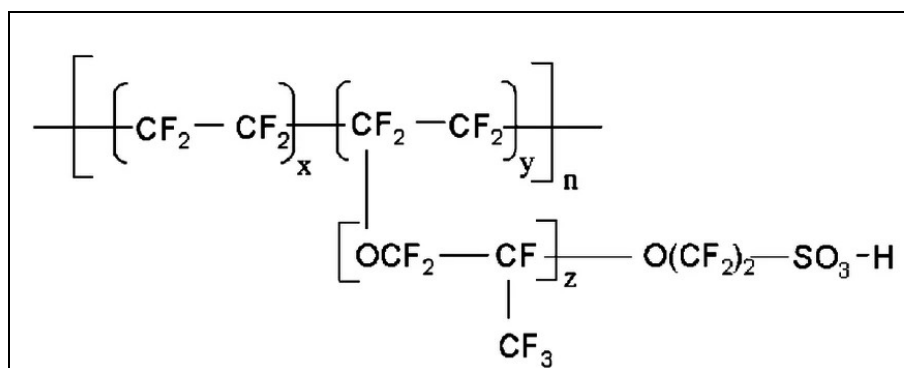


Figure 2 : Nafion molecular structure [3]

Measurements on Nafion 117 demonstrated that water uptake and transport properties depend on membrane water content [4]. Consequently, hydration is a key consideration when modifying Nafion for low humidity operation. At elevated temperatures and low relative humidity, membrane dehydration reduces proton conductivity [5], while thermal and humidity cycling induces dimensional changes that may compromise membrane integrity. High methanol crossover also limits its performance in direct methanol fuel cells [6]. Besides, its relatively high cost and susceptibility to radical-induced chemical degradation further motivate the development of Modified Nafion membranes[7]. Incorporating suitable nanofillers has therefore been investigated to improve water

retention and proton transport. Generally, nanofillers can be grouped into 6 main categories as shown in Figure 3.

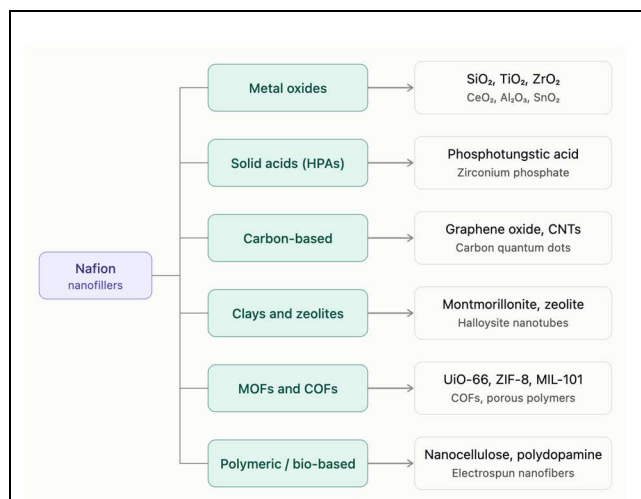


Figure 3 : Nafion nanofillers categories

They fillers can be classified into six groups: metal oxides, solid acids, carbon based, clay and zeolites, metal and organic frameworks (MOFs) and covalent organic frameworks (COFs) and polymeric and biobased materials. Each group can modify Nafion through distinct, although often overlapping mechanisms as indicated in Table 1.

Table 1 : Main group of nanofiller with its function

Group	Function
Metal Oxide (SiO ₂ , TiO ₂ , ZrO ₂ , CeO ₂)	hold water at high temperature and low humidity. They also improve mechanical strength. CeO ₂ additionally scavenges radicals, which slows chemical degradation.
Solid acids heteropolyacids	as proton conductors that increase the conductivity
Carbon based (graphene oxide, Carbon Nanotube)	add mechanical strength, and sulfonated forms add proton-conducting sites.
Clays and zeolites	lower fuel crossover (such as methanol) and help retain water.
MOFs and COFs	have tunable pores and functional groups that create proton-conduction pathways.
Polymeric / bio-based (nanocellulose, polydopamine, electrospun nanofibers)	improve dimensional stability and add interfacial compatibility

One widely studied approach is to incorporate ceramic, polymeric or carbon-based fillers into the Nafion matrix. These composite membranes aim to improve water retention, proton transport and durability while retaining the useful properties of the original polymer [8]. Common fillers include hydrophilic oxides such as SiO₂, TiO₂ and ZrO₂, radical-scavenging ceria, and functionalised graphene oxide and carbon nanotubes. Sulphonation can introduce additional acid groups that support proton conduction, although the benefits depend on filler loading, dispersion and interactions with the polymer [9, 10].

Recent studies illustrate the potential of these modifications. Quan et al. (2026) reported that a sol-gel-prepared Nafion/SiO₂ composite achieved a proton conductivity of 61.9 mS·cm⁻¹ at 120 °C and

50% relative humidity, with open-circuit voltage and peak power density exceeding those of unmodified Nafion by 11.2% and 8.9%, respectively [11]. In a separate study, Segale et al. (2024) reported an ion-exchange capacity of $1.81 \text{ meq}\cdot\text{g}^{-1}$ for a Nafion silica composite, compared with $0.93 \text{ meq}\cdot\text{g}^{-1}$ for pristine Nafion [12]. Waste-derived materials offer another direction for investigation. Pratama et al. (2025) examined palm oil fuel ash-derived silica in chitosan membranes, providing a basis for exploring similar fillers in Nafion rather than direct evidence of their performance in a Nafion matrix [13]. This review therefore examines the principal filler strategies Nafion nanocomposite membranes.

2.0 NAFION STRUCTURE AND PROTON TRANSPORT

Nafion structure and proton transport mechanism as illustrated in Figure 4. Figure 4(a) shows chemical structure of nafion consisting of a hydrophobic fluorocarbon backbone and hydrophilic sulfonic acid groups.

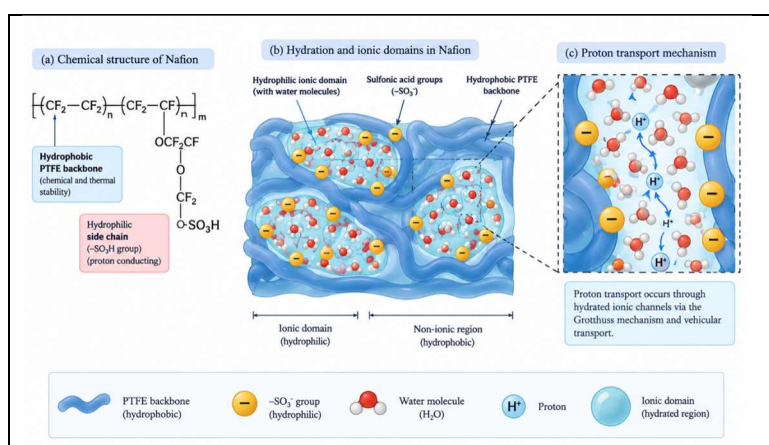


Figure 4 : Schematic representation of Nafion structure, hydration and proton transport

The chemical structure allows the polymer to form distinct hydrophobic and hydrophilic regions as indicated in Figure 4 (b). Figure 4 (c) shows the hydrophilic regions contain sulfonic acid groups that attract water and form pathways that facilitate proton transport. This phase separated morphology is a key reason for Nafion's widespread use as a Proton Exchange Membrane[14]. The morphology of nafion is strongly related to its water uptake and proton conductivity [15]. When water enters the membrane, the sulfonic acid groups become hydrated and the hydrophilic ionic domains expand. Hydration of these domain facilitates proton transport through vehicular and proton transport mechanism. Consequently, the amount and distribution of water within nafion are important factors controlling its electrochemical performance [14]

3.0 PRINCIPAL FILLER STRATEGIES FOR NAFION NANOCOMPOSITE MEMBRANE

Filler incorporation offers a way to modify Nafion's hydration, proton transport and structural stability. The effectiveness of this approach depends on the filler's surface chemistry, loading and distribution within the membrane. Hydrophilic fillers can retain water near the sulfonic acid groups, while acid-functionalised additives can introduce additional sites for proton transport. Other fillers provide mechanical reinforcement or protection against radical-induced degradation. These functions are complementary, but improvements in one property may come at the expense of another. Filler selection should therefore consider the balance between conductivity, dimensional stability and durability under the intended operating conditions.

3.1 Metal Oxide Fillers

Metal oxide fillers offer a practical approach to improving water retention and mitigating hydration-dependent performance losses in Nafion membranes. Metal Oxides such as SiO_2 , TiO_2 and ZrO_2 have been investigated primarily for improving water retention and maintaining proton transport at elevated temperatures and reduced relative humidity. Their contribution depends on the oxide composition and its interaction with the ionomer, rather than simply on the presence of a hydrophilic second phase. The preparation method strongly influences particle dispersion and the resulting polymer–filler interface. Two principal approaches are used: in situ sol–gel synthesis, in which oxide particles form within a Nafion membrane or dispersion, and solution blending followed by recasting, in which separately prepared particles are incorporated into the ionomer.

Jalani et al (2005) demonstrated that sol–gel-derived Nafion/ MO_2 composites, where M represents Si, Ti or Zr, exhibited greater water sorption than unmodified Nafion at 90 and 120 °C [16]. Li et al (2010) obtained well-dispersed SiO_2 and ZrO_2 nanoparticles with diameters of approximately 5 nm using a self-assembly-assisted sol–gel process [17]. Under the conditions examined, particle incorporation did not significantly alter Nafion crystallinity or structure. The authors attributed the restricted particle growth to the association of Nafion molecules with the oxide surfaces. These observations emphasise the importance of precursor hydrolysis, condensation and interfacial stabilisation in obtaining homogeneous nanocomposite membranes.

Amjadi et al (2012) prepared Nafion/ SiO_2 membranes through in situ sol–gel synthesis and showed that processing conditions influenced particle leaching, highlighting the need to consider filler retention alongside initial electrochemical performance [18]. More broadly, mixed oxides provide opportunities to adjust membrane behaviour through compositional control. Park et al. (2008) investigated recast Nafion membranes containing ZrO_2 – SiO_2 particles and found that all examined composites exhibited higher water uptake than the unmodified membrane [19]. Despite these benefits, oxide incorporation requires careful consideration of stability across the complete membrane–electrode assembly. Ceria, for example, can protect the membrane while introducing adverse effects elsewhere in the cell. Yuk et al (2023) reported that Ce^{4+} released from CeO_2 contributed to Pt/C catalyst degradation and reduced power performance [20].

3.2 Solid Acid Nanofillers

Building on the hydration benefits of oxide fillers, solid acid additives introduce acidic sites that can contribute to proton transport within Nafion composites. Representative materials include heteropolyacids, zirconium phosphate and sulfated metal oxides. Their effectiveness depends on the accessibility of these sites, membrane hydration and the stability of the inorganic phase within the polymer. Ramani et al. (2004) investigated Nafion composites containing phosphotungstic, silicotungstic, phosphomolybdic and silicomolybdic acids for operation at 120 °C and 35% relative humidity [21]. The composites exhibited proton conductivities of approximately 0.015 S cm^{-1} under these conditions.

Heteropolyacids such as phosphotungstic acid (PWA), are promising proton conducting additives because their strong acidity. However, their water can compromise long term retention within the membrane [22]. Addressing this limitation, Xu et al (2021) stabilised PWA within Nafion through targeted silica fixation, using PWA to catalyse the in situ hydrolysis of the silica precursor [23]. The resulting PWA/silica/Nafion membrane exhibited a proton conductivity of 0.058 S cm^{-1} at 110 °C and 60% relative humidity, approximately 2.4 times that of the pristine Nafion reference. The authors attributed this improvement to the combined contributions of proton-conducting PWA and water-retaining silica within the ionic domains.

An alternative immobilisation strategy uses porous organic frameworks to retain heteropolyacids and organise proton-transport pathways. Zhai et al (2023) chemically bonded PWA within a covalent organic framework to produce a water-insoluble hybrid filler for Nafion [24]. At a loading of 4 wt%, the composite achieved a proton conductivity of 0.061 S cm^{-1} at 25 °C and 55%

relative humidity. Its membrane-electrode assembly also delivered a peak power density of 724 mW cm^{-2} at 80°C and 50% relative humidity, representing a 138% increase over the recast Nafion reference. More recently, Lu et al (2026) investigated PWA-functionalised CeO_2 incorporated into Nafion, extending the concept towards fillers that combine acidic functionality with improved chemical stability. These studies illustrate a progression from direct acid incorporation towards hybrid architectures designed to integrate acid retention, proton transport and membrane durability.

3.3 Polymeric and Biobased Nanofillers

Organic and polymeric additives offer opportunities to adjust hydration and proton transport through their functional groups. Poly(vinyl alcohol) (PVA) is a widely investigated blending component because its hydroxyl groups promote hydrogen bonding and influence water and methanol transport. Ng et al. (2023) reported a 40.33% reduction in methanol permeability relative to recast Nafion, illustrating the potential of PVA to improve fuel rejection [26]. Nevertheless, unmodified PVA lacks strongly acidic proton-exchange groups, and the balance between barrier performance and conductivity remains formulation dependent.

Cellulose-based fillers provide a complementary means of modifying hydration and mechanical response. Hambardzumyan et al. (2022) compared Nafion composites containing ramie- and tunicate-derived cellulose nanocrystals (CNCs), showing that filler content and aspect ratio affected water uptake, anisotropic swelling and mechanical properties [27]. Increasing CNC content to 10 wt% increased water uptake and thickness swelling while reducing in-plane swelling; a 5 wt% tunicate-CNC loading provided the best mechanical compromise. Biobased functionalization also extends to lignin. Heo et al (2023) demonstrated enhanced proton transport in synthesized zwitterion-substituted lignin/Nafion composites relative to composites containing unmodified lignin, together with improved oxidative stability for a selected formulation compared with pure Nafion [28]. This comparison indicates that lignin functionality and loading are central to performance; improvements cannot be attributed solely to its renewable origin. Moreover, agricultural waste also offers another cellulose-based route. Talarico et al (2025) incorporated lemon-waste nanocellulose into Nafion from a hydroalcoholic dispersion, obtaining the highest conductivity at 10 wt% loading and improved flexibility and ductility [29]. Conductivity exceeded that of pristine Nafion from 50°C under the reported measurement conditions. Thus, the suitability of biobased filler should be evaluated based on its functional contribution, processing compatibility and long term membrane performance rather than renewable origin alone.

3.4 Carbon Based Nanofillers

Carbon-based modifications of Nafion include dispersed nanofillers, surface coatings and layered composite architectures based on Graphene oxide (GO), graphene and carbon nanotubes (CNTs). Ceballos-Alvarez et al (2025) showed that annealing promoted interaction between deposited GO and commercial Nafion 115, supporting improvements in mechanical response [30]. Ruhkopf et al (2023) deposited electrochemically exfoliated graphene onto Nafion membranes, forming a diffusion barrier that reduced hydrogen permeability by up to a factor of 29 at 80°C and 65% relative humidity [31]. These findings indicate that graphene-based barrier layers can effectively suppress gas crossover.

CNT-based fillers combine an elongated reinforcing structure with chemically tunable surfaces. Yin et al (2018) demonstrated that sulfonated CNTs modified the evolution of ionic-water clusters in Nafion and promoted hydrated pathways connecting otherwise isolated clusters under low-humidity conditions [32]. Hybridization with hygroscopic oxides offers another approach. Nicotera et al (2023) grew TiO_2 nanoparticles on multiwalled CNTs and incorporated the hybrid into Nafion [33]. At an optimized 3 wt% loading, the membrane supported a peak power density of 307.7 mW cm^{-2} in an H_2 /air fuel cell at 120°C and 30% relative humidity, compared with 92.2 mW cm^{-2} for the Nafion 212

reference. The authors associated the improvement with enhanced water retention and transport. Higher loading impaired water mobility, highlighting the need to optimize dispersion and concentration.

3.5 Clays and Zeolites Nanofillers

Clay and zeolite fillers provide complementary routes to controlling fuel crossover, hydration and mechanical response in Nafion membranes. Early Nafion/ Layered montmorillonite (MMT) studies established the potential of layered barriers to suppress methanol transport as reported by Song et al (2004). They reported reduced methanol crossover in Nafion/organo-MMT composites at a clay loading as low as 1 wt% [34]. Beauger et al (2013) demonstrated that sepiolite incorporation increased water uptake and improved the mechanical properties of Nafion; sulfonation increased the ion-exchange capacity of the filler and composite membranes. Their optimized composite achieved approximately 50% higher fuel-cell power at 100 °C and 50% relative humidity than the pure Nafion reference [35].

Recent sepiolite research combines mineral scaffolds with confined ionic liquids and controlled fiber organization. Ling et al. prepared a sepiolite-supported ionogel using 1-butyl-3-methylimidazolium trifluoromethanesulfonate and supercritical CO₂, then incorporated it into electrospun Nafion composites [36]. The authors attributed the enhanced performance to axial alignment of the ionogel within the Nafion nanofibers. The selected composite delivered a peak power density of 779 mW cm⁻² at 60 °C and 80% relative humidity.

Zeolite-containing Nafion membranes similarly require a balance between diffusion resistance and proton conduction. Chen et al (2006) formed acid-functionalized zeolite Beta within Nafion by in situ hydrothermal crystallization found the composite exhibited approximately 40% lower methanol permeability [37]. Zanchet et al (2021) formed zeolite nanocomposite with combined HZSM-5 and an imidazolium triflate ionic liquid with Nafion. He reported the membrane improved water absorption and proton conductivity while retaining thermal stability, together with improved PEMFC performance at 100 °C under low-humidity conditions [38].

3.6 MOF and COF Nanofillers

Metal-organic frameworks (MOFs) and covalent organic frameworks (COFs) offer chemically tunable porous fillers for modifying proton transport in Nafion. For instance, Wang et al (2021) reported that a membrane containing 5% Zn-MOF reached 7.29×10^{-3} S cm⁻¹ at 80 °C and 58% relative humidity (RH), approximately 1.87 times the conductivity of pure Nafion [39]. Separately, Shao et al (2023) developed ZUT-COF-SO₃H/Nafion membranes, associating their enhanced transport with COF nanochannels, sulfonic-acid functionality and water retention; the reported apparent activation energy was 0.086 eV [40]. More recently, Zhang et al (2025) prepared NUS-9/Nafion composites through incorporation of interfacially synthesized sulfonated nanosheets. Improved hydration, ionic-cluster connectivity and mechanical response accompanied a maximum power density of 1.024 W cm⁻² at 0.5 wt% loading, approximately 80% above pristine Nafion in their cell tests. The study also reported improved durability [41]. Collectively, these studies suggest that performance MOF/COF containing Nafion membranes arise from the combination of controlled porosity, functional proton-conducting groups and enhanced water retention. However, the reported conductivity and power density improvements were obtained under different membrane compositions and operating conditions, preventing direct ranking of the fillers.

4.0 Future direction of Nafion Composite Membrane

Future development of Nafion composite membranes should prioritize the rational design of hybrid fillers that balance proton transport, crossover suppression and operational durability. Combining water-retaining and barrier-forming materials with proton-conducting, reinforcing or radical-scavenging components offers a promising route to addressing these interconnected requirements. The benefits of such combinations depend on interfacial compatibility, filler dispersion and the continuity of proton-conduction pathways, making controlled orientation and membrane architecture as important as filler composition. Computational screening and machine learning could support material selection, while experimental validation must establish sustained performance under application-specific conditions. Key priorities include maintaining proton conductivity at elevated temperatures and low relative humidity, limiting fuel and gas crossover, and resisting dimensional changes and chemical degradation during prolonged operation. Renewable fillers, reduced perfluorosulfonic acid content and greener processing may also lower material consumption and environmental burdens, provided that these benefits are demonstrated without compromising performance. Progress toward commercially viable membranes will therefore require integrated optimization of transport, stability, manufacturability and cost, supported by consistent characterization and long-term testing in fuel cells and water electrolyzers.

5.0 Conclusion

This review highlights the potential of six major nanofiller categories to improve the performance of Nafion composite membranes through enhanced water retention, proton conductivity, mechanical stability, and resistance to fuel crossover and chemical degradation. Each category offers distinct advantages, but its effectiveness depends on filler properties, loading, dispersion, and compatibility with the Nafion matrix. The reviewed studies demonstrate that no single nanofiller can address all membrane limitations; therefore, selecting suitable materials requires balancing proton transport, dimensional stability, and durability. Overall, nanofiller incorporation provides a versatile approach to developing high-performance Nafion membranes for proton exchange membrane fuel cells and water electrolyzers.

Author Contribution

S. Abdullah M. Mohd: Conceptualization, investigation, visualisation, writing and editing. N.S.N.N.S.Alam.: Investigation, writing, and editing. N.Hashim : Conceptualization, investigation. Hassan. L.S: Conceptualization, investigation. N.A.M. Sobri : Conceptualization, investigation

Conflict of Interest

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

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