*J. Applied Membrane Science & Technology*, Vol. 29, No. 2, August 2025, 193–208


# Effect of Hot Pressing Parameters on the Performance of Graphene Oxide Speek Membrane Electrode Assembly for Proton Exchange Membrane Fuel Cell Application

Kenny Looi Soon Ken[a,b], J. Jaafar[a,b], N. S. Suhaimin[a,b]*

[a]School of Chemical and Energy Engineering, Faculty of Engineering, Universiti Teknologi Malaysia, 81310 UTM Johor Bahru, Johor, Malaysia

[b]Advanced Membrane Technology Research Centre (AMTEC), Universiti Teknologi Malaysia, 81310 UTM Johor Bahru, Johor, Malaysia

Submitted: 28/5/2025. Revised edition: 28/7/2025. Accepted: 28/7/2025. Available online: 1/8/2025

## ABSTRACT

Fuel cells have garnered significant research attention over the past decades due to their versatility across portable, transportation, and stationary applications. Among various types, the Proton Exchange Membrane Fuel Cell (PEMFC) stands out as the most promising due to its ability to generate clean energy using hydrogen and oxygen without carbon dioxide emissions. Nafion, the commercial proton-conducting membrane used in PEMFCs, offers excellent proton conductivity along with high chemical and thermal stability. However, its high cost and limited operational temperature range (up to 80 °C) poses significant challenges for practical applications. Therefore, this study explores a cost-effective alternative nanocomposite membrane based on sulfonated polyether ether ketone (SPEEK) incorporated with graphene oxide (GO) as a carbonaceous filler. GO was synthesized via a modified Hummer's method and integrated into the SPEEK matrix. The resulting nanocomposite membrane was characterized using Scanning Electron Microscopy (SEM) for morphological analysis and Fourier Transform Infrared (FTIR) spectroscopy to identify functional groups. The Membrane Electrode Assembly (MEA) was optimized using hot-pressing at 110 °C under 5 tons for 4 minutes, achieving a peak power output of 184.02 mW. The results demonstrate that the inclusion of GO enhances the performance of the PEM, positioning GO-SPEEK as a promising and cost-effective substitute for Nafion in PEMFC applications.

*Keywords*: Proton exchange membrane fuel cell (PEMFC), polymer electrolyte membrane (PEM), sulfonated poly(ether ether ketone) (SPEEK), membrane electrode assembly (MEA), graphene oxide (GO), hot-press parameter

## 1.0 INTRODUCTION

Proton exchange membrane fuel cell (PEMFC) is the most convincing energy conversion device for stationary and mobile applications due to its high energy efficiency and low operation temperature. Hydrogen is one of the fuels used by proton exchange membrane fuel cell (PEMFC) and it is proven to generate energy 5 times higher than conventional energy [1]. Despite that, hydrogen is greatly suitable as a clean energy source and reduce carbon dioxide emissions. Nonetheless, the difference between fuel cells and batteries is fuel cell can run continuously if the reactants are constantly refilled from a storage device and not fully depleted in the electrochemical reactions [2]. Nafion is currently the popular proton exchange membrane in PEMFC. The membrane is manufactured with fluorinated

* Corresponding to: N. S. Suhaimin (email: sariyan@utm.my)
DOI: https://doi.org/10.11113/jamst.v29n2.320

polymer which hydrophobic polytetrafluoroethylene (PTFE) also known as Teflon, attached to the structured backbone and linked with the hydrophilic sulfonic acid (-$SO_3H$) [3]. Hydrophobic PTFE gives great thermal and chemical properties while hydrophilic sulfonic acid makes Nafion an excellent proton-conducting membrane respectively [4]. Most importantly, Nafion is the first PEM that introduced the major breakthrough in the fuel cell history because it allows PEMFC to operate without the presence of acid electrolyte and provide higher fuel cell performances.

While Nafion able to provide higher fuel cell performances [5], its complex manufactured structure is very costly and its conductivity only dependent on the availability of water [24]. Moreover, there are some setbacks in Nafion that prohibit its ability to reach the highest levels. For example, Nafion is only optimum for operating at temperatures within 80 °C compared to perfluorinated ionomer (PFI) which commonly used in high-temperature proton exchange membrane (PEM) fuel cells [25]. Hence, it is necessary to find an alternative PEM with higher thermal stability and cheaper manufacture cost.

Sulfonated poly(ether ether ketone)s (SPEEK)s are aromatic main-chain polymers undergoes sulfonation [6], [7]. SPEEK is the most studied polymer for the PEMs because its chemical structure alone gives better mechanical strength, good thermal stability and cheap to manufacture the polymer. Furthermore, it is proven that SPEEK membrane can be enhanced with nanofiber composite fabrication to obtain higher proton conductivity and stable chemical structure [8]. Incorporation of carbonaceous fillers, GO in SPEEK matrix will improves the properties of the nanocomposite membrane, such as thermal stability, mechanical stability and proton conductivity due to the presence of oxygen functional groups [9]. In addition, these excellent properties are attributed from the 2D nanostructures of graphene which the lateral size eases the flow of electrolyte ions [10], [11]. Although Hummer's method was first developed in 1958, it is still one of the most commonly used methods for synthesizing graphene oxide (GO) from graphite. While newer techniques such as electrochemical exfoliation, microwave-assisted synthesis, and green methods [26], [27], [28], [29], [30] have emerged, Hummer's method remains widely used because it is simple, efficient, and scalable. Modified versions of the method have also been introduced to reduce harmful byproducts and improve oxidation efficiency such as by removing sodium nitrate or using safer oxidizing agents [9], [31]. These improvements have made the method more environmentally friendly while maintaining its effectiveness. Owing to its reliability and the high-quality GO produced, the method is still relevant today, especially for studies focusing on the structure, properties, and further functionalization of GO.

Hot pressing plays a significant role in the fabrication and performance optimization of membrane electrode assemblies (MEAs), especially in systems employing graphene oxide (GO)-based and sulfonated poly(ether ether ketone) (SPEEK) composite membranes for proton exchange membrane fuel cell (PEMFC) applications. The process involves the simultaneous application of heat and pressure, which directly influences the microstructural characteristics and interfacial integrity of the membrane. Specifically, hot pressing promotes densification of the GO-SPEEK membrane by reducing interlayer spacing and enhancing interfacial contact between the membrane and

electrode layers, leading to improved proton conductivity and reduced interfacial resistance. It also strengthens mechanical integrity by enhancing interlayer interactions, such as van der Waals forces and hydrogen bonding, which are crucial for maintaining membrane stability during fuel cell operation [32]. Additionally, elevated temperatures during hot pressing may induce partial rearrangement or reduction of oxygen-containing functional groups in graphene oxide, potentially improving membrane hydrophobicity balance and thermal stability [33]. Moreover, by proper tuning of hot pressing parameters, such as temperature, pressure, and duration, it would ensure strong adhesion at the electrode–membrane interface, thus minimizing delamination and improving long-term performance of the MEA [33]. Undoubtedly, the optimization of hot pressing conditions is essential for achieving high power output, stable operation, and durable MEA performance in PEM fuel cell systems

In this study, carbonaceous filler, graphene oxide (GO) is successfully prepared with modified Hummer's method and crosslinked into sulfonated polyether ether ketone (SPEEK) membrane. Morphological structures of nanocomposite membrane is identified with SEM and the functional groups of GO SPEEK is determined using FTIR spectroscopy. Optimum hot-pressing parameters of GO SPEEK MEA is investigated at several variables in external loop single cell and the performance of the MEAs are evaluated from IV-polarization curves.

## 2.0 METHODS

### 2.1 Materials

Poly(ether ether ketone) (PEEK) polymer in powder form was provided by Victrex US Inc. Ltd, 1-Methyl-2-pyrrolidone (NMP) with purity (GC) ≥ 99.9% was obtained from Merck Co., Germany and Sulfuric acid (H2SO4, 95-97%) was supplied by QReC, Graphite powder with a purity of 99.99 wt% was obtained from Sigma Aldrich, the electrodes (platinum-ruthenium (40%) on carbon cloth and 40% platinum (40%) on carbon cloth) were acquired from Fuel cell earth.

### 2.2 Fabrication of Flat Sheet SPEEK Membrane

#### *2.2.1 Sulfonation of PEEK*

SPEEK is prepared by sulfonating the PEEK powder using concentrated sulfuric acid under controlled temperature and stirring. 1 L of sulfuric acid is poured into a 2 L beaker that has been placed on a hot plate.1 L of sulfuric acid is poured into a 2 L beaker that has been placed on a hot plate. A thermometer and an automatic stirrer are placed within the beaker before closing with the aluminium foil. After that, 50 g of PEEK powder that has been dried overnight in an oven is added steadily into the beaker as the solution is mixed clearly at room temperature for an hour. After an hour, the temperature is increased to 63°C to heat up the mixture for 3 hours. After 3 hours, the mixture is precipitated in a water strainer filled with ice. Then the SPEEK precipitation is washed until the precipitation has been neutralized. The neutralized SPEEK precipitation is placed in an aluminium foil shaped as a cup and dried overnight at 80 °C in an oven.

#### *2.2.2 SPEEK Solution Formulation and Membrane Casting*

To produce a 10 wt.% of SPEEK dope solution, 10 g of dried SPEEK precipitation is added to 90 g of 1-Methyl-2-pyrrolidone (NMP) with a

magnetic stirrer and left overnight to dissolve completely. Next, 10 mL of dope solution is added into a 10 cm x 10 cm mold and it is placed into the oven overnight. On the next day, the membrane is peeled off from the mold and stored for MEA assembly.

### 2.3 Synthesis of Graphene Oxide (GO)

Graphene oxide can be prepared by modified Hummer's method [9] using strong oxidizers such as potassium permanganate ($KMnO_4$) and sodium nitrate ($NaNO_3$) together with concentrated sulfuric acid ($H_2SO_4$) intercalating agent. 2 g of graphite powder is slowly added into 50 ml of concentrated sulfuric acid in ice bath. 6 g of potassium permanganate ($KMnO_4$) and 2 g of sodium nitrate ($NaNO_3$) is then gradually added into the solution under continuous stirring for 1 hour. The mixture's temperature will be maintained around 35℃ before the reaction terminated. The reaction is terminated by adding 350 ml of distilled water and 10 ml of hydrogen peroxide (30% $H_2O_2$). The mixture is then left overnight. The mixture then is washed several times using 5% hydrochloric acid (HCl) to remove residual metal ions such as $Mn^{2+}$ and $Na^{+}$ formed during the oxidation reaction. This washing step is important to ensure that the GO is free from impurities that may affect membrane performance. After washing, the sample is rinsed with deionized water until the pH becomes neutral. Finally, the filtered GO is dried in the oven at 80 ℃ for a day to obtain the dried GO powder. The GO produced shows good blackish-brown colour.

### 2.4 Fabrication of Flat Sheet GO SPEEK Membrane

Fabrication of GO SPEEK membrane begins by dissolving 0.25 g of dried GO in 10 ml of 1-methyl-2-pyrrolidone (NMP) while dissolving 10 g of dried SPEEK in 90 ml of NMP for a day. After that, the two dope solutions will be mixed and proceeded to heat to 100℃ with continuous stirring for an hour before casting. The mixture will be casted on a petri dish and dried in the oven overnight at 80℃.

### 2.5 Characterization of Graphene Oxide (GO), SPEEK and GOSPEEK

The surface morphology analysis of the composite membrane was observed using a scanning electron microscopy (SEM-TM 3000) at an accelerating voltage of 15 kV. The membrane samples were prepared by submerged it into liquid nitrogen for a few seconds before passed through for gold sputtering. The Fourier transform infrared (FTIR) spectroscopy was used to obtain some information on the functional groups present in the membranes and inorganic filler.

### 2.6 Water Uptake

To study the hydrophilicity of the membrane, the water uptake (WU) of the membranes were carried out through measuring the changes in the weight of the samples in the dry and wet conditions. After weighing the weight ($W_{dry}$), the dry membrane was soaked in deionized water for 24 h at room temperature and then recorded the weight ($W_{wet}$) of the wet sample. WU of the membrane are calculated according to the following equation:

$$\text{Water uptake} = \frac{W_{wet} - W_{dry}}{W_{dry}} x\, 100. \quad (1)$$

### 2.7 Preparation of Membrane Electrode Assembly (MEA)

Fabrication of membrane electrode assembly (MEA) was carried out by hot-pressing the membrane between the

anode and cathode electrodes under three important hot-pressing parameters including, time, pressure and temperature. The membrane was cut into 3.0 cm x 3.0 cm while the electrodes were cut into 1.5 cm x 1.5 cm. The hot-pressing procedures was conducted at various time, pressure and temperature as presented in Table 1.

**Table 1** Hot pressing parameters on the GO SPEEK MEA

| Hot Pressing Parameters | Range |
|---|---|
| Temperature (°C) | 90 to 120 |
| Time (min) | 4 to 7 |
| Pressure (ton) | 5 to 7 |

### 2.8 Single PEMFC Performance Testing

The performance of GOSPEEK MEA as a single-cell PEMFC is analyzed by using an external looped fuel cell analyzer test and it is compared with SPEEK. The sample GO SPEEK MEA is placed between the anode and the cathode plates. Hydrogen ($H_2$) outlet is flowing into the anode at 75 ml/min and the cathode side is supplied with oxygen ($O_2$) by directing a fan at the MEA under ambient temperature. Then, the sample MEAs are compared to determine the highest maximum power density and achieve the optimum parameters for the hotpressing technique based on the plotted I-V polarization and power density curves.

## 3.0 RESULTS AND DISCUSSION

### 3.1 Morphological Structural of GO and SPEEK-based Membrane

Figure 1 (a) below shows the SEM images of graphene oxide (GO) at 2000x magnification. Based on the image, some aggregates are seen on the surface of the nanosheet GO due to the stacking between neighboring individual GO sheets. This can confirm that the GO is synthesized from modified Hummer's method [37][38]. In comparison of Figure 1 (b and c), the surface of SPEEK and GO SPEEK membranes look dense and smooth which indicate that SPEEK polymer and GO exhibit excellent dispersibility in 1-methyl-2pyrrolidone (NMP). The cross-section image of SPEEK and GO SPEEK membranes are depicted in Figure 1(d and e), respectively. Parent SPEEK membrane shows a smooth surface on the cross-section indicating the uniformity and well-dense structure. Meanwhile, Figure 1 (e) shows that the addition of GO in SPEEK has disrupted the continuous phase of SPEEK matrix and exhibited some sites with crumples (in red circle) proven the existence of GO nanoparticles. The presence of wrinkled or crumpled features observed in SEM has been similarly reported in GO-containing membranes by other studies [41][42]. As expected, GO SPEEK membrane was successfully fabricated in dense structure due to strong interaction between GO and SPEEK chains. While such aggregation can potentially reduce the overall surface area and limit the accessibility of individual nanoscale active sites, it does not completely diminish the functional advantages of GO. In fact, aggregated GO structures have been shown to retain significant activity due to their layered morphology and the abundance of oxygen-containing functional groups, which facilitate water uptake and proton conduction [34], [35], [36]. Moreover, these clusters may enhance mechanical reinforcement and create continuous, interconnected pathways that support efficient proton transport across the membrane, which reflected in the

improved electrochemical performance of the GO SPEEK membrane observed in this study.

### 3.2 Water Uptake

Water uptake of SPEEK membrane represents the availability of the sulfonic acid group of SPEEK to attach with water molecules. It is proven that the percentage of water uptake depends on the degree of sulfonation in the SPEEK [6]. At a sulfonation degree of 67%, a sulfonic acid group of SPEEK can attach to 4.5 water molecules. Water molecules absorbed will act as a medium for the transport of protons in the SPEEK membrane. Nevertheless, crosslinking of composite membrane will affect the water uptake capabilities, either improving or limiting the flow of water molecules. Based on Figure 2, it can be proven that the GO SPEEK membrane showed higher water uptake compared to the pristine SPEEK membrane. At 30 °C, the water uptake of pristine SPEEK was 57.63%, while GO SPEEK showed 78.24%. As the temperature increased to 60 °C and 90°C, the water uptake values increased to 91.08% and 118.36% for SPEEK, while 115.37% and 168.15% for GO SPEEK. This shows that the presence of GO in the membrane improves hydrophilicity due to the hydroxyl (–OH) functional groups that attract more water molecules [12]. Therefore, GO SPEEK membrane has better water absorption capability, which helps to improve proton conductivity at higher temperature. The reason is the hydrophilic groups (OH-) in the structure of GO enable the nanosheet to consist high attraction towards the water resulting increase the water uptake capacity [12]. Hence, crosslinking of composite membrane as shown in the figure is viable to increase the water uptake and improve the proton conductivity of the membranes.

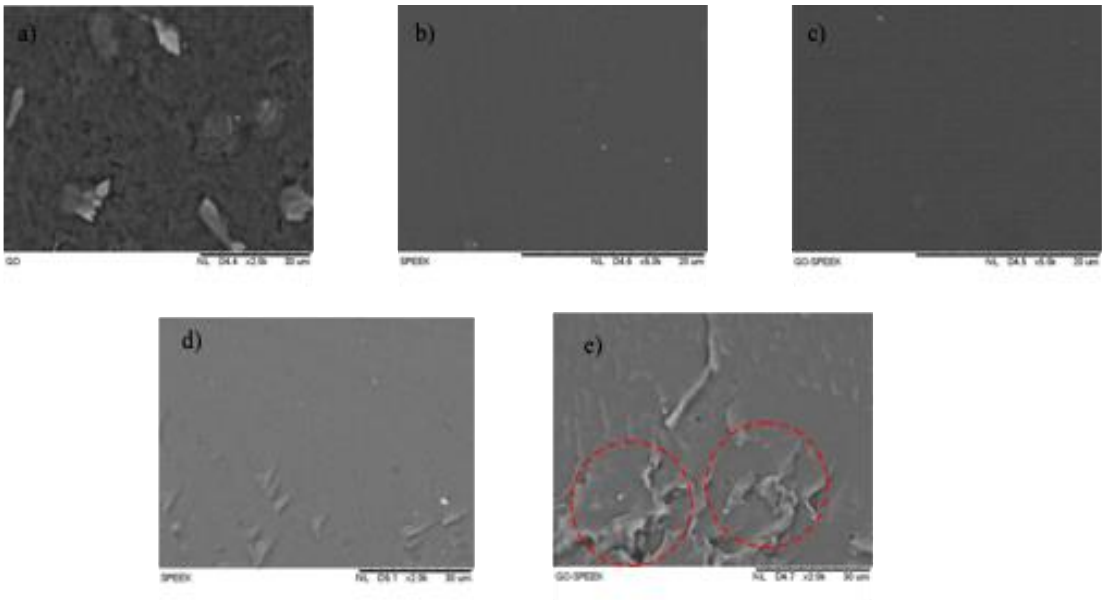


**Figure 1** SEM images of (a) graphene oxide (GO), (b) surface of SPEEK membrane, (c) surface of GO SPEEK membrane, (d) cross-section of SPEEK membrane, and (e) cross-section of GO SPEEK membrane

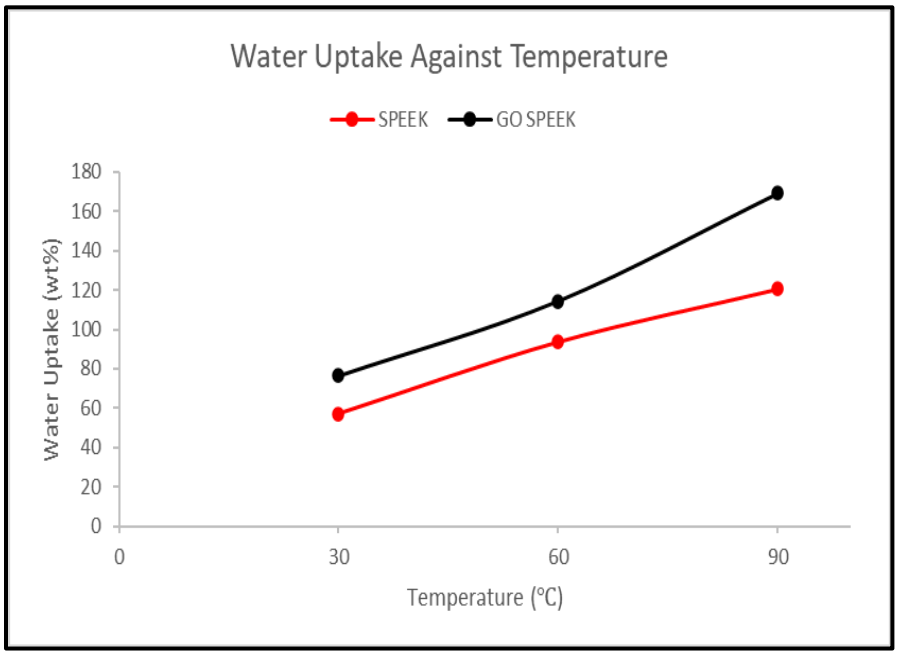


**Figure 2** Water uptake of SPEEK and GO SPEEK membranes prepared at several temperatures

### 3.3 Fourier Transform Infrared (FTIR) Analysis

The structural properties of SPEEK and GO SPEEK were characterized using FTIR spectroscopy, as shown in Figure 3. Both spectra exhibit a broad absorption band in the range of 3500–2000 $cm^{-1}$, which is attributed to the O–H stretching vibrations. This peak originates from the hydroxyl groups present in both the sulfonated SPEEK polymer and the oxygen-containing functional groups of graphene oxide (GO), such as hydroxyl and carboxyl moieties introduced during oxidation. The presence of these hydrophilic groups is critical for membrane hydration, as they facilitate the absorption and retention of water molecules within the membrane matrix. This in turn promotes the formation of hydrogen-bonded networks essential for proton conduction via the Grotthuss mechanism [34]. Therefore, the intensity and broadness of the O–H peak not only confirm successful sulfonation and GO integration but also underscore their functional relevance to the enhanced proton conductivity and fuel cell performance of the GO-SPEEK membrane. The characteristic peak at 1644.61 $cm^{-1}$ represents the carbonyl functional group (C=O) vibrations followed by the arising epoxide group (C-O) at 1594.28 $cm^{-1}$. These two peaks represent oxygen functional groups which confirmed that the graphite was oxidized into graphene oxide during the modified Hummer's method [13]. The peak at approximately 1217 $cm^{-1}$ is corresponded to the aromatic C-O-C. Most importantly, both GOSPEEK and SPEEK showed similar characteristics bands at approximately 1077 and 1021 $cm^{-1}$ is attributed to the the sulfonic groups (-$SO_3H$) [14]. Nevertheless, the increase of electrostatic force between GO and SPEEK causes the peak intensity of sulfonic groups to decreased compared to pristine SPEEK membrane [15]. Figure 4 below indicates the chemical structure of GOSPEEK.

### 3.4 Optimum Hot-Pressing Parameters of GOSPEEK Membrane Electrodes Assembly (MEA)

When heat and pressure are applied during hot pressing, the membrane structure becomes more compact as the polymer chains are packed closely together. This condition reduces the amount of water that can be absorbed into the membrane due to the decrease in porosity and tighter chain arrangement [32], [39]. However, it still able to retain enough water to support proton conduction [39], [40].

Moreover, hot pressing improves the membrane density and mechanical strength. The alignment of polymer chains and the presence of GO sheets make the membrane structure stronger and more stable [32], owing to better interaction between GO and SPEEK. Even though the membrane absorbs less water after hot pressing, the proton conductivity remains high because the GO contains hydrophilic groups such as –OH, –COOH and –$SO_3H$. These groups continue to help with proton transfer across the membrane [14].Therefore, the optimum hot-pressing parameters is highly desirable because it is dominantly affecting the overall performance of the GOSPEEK MEA in PEMFCs [16]. Based on Figure 5 (a and b), a range of hot-pressing temperatures from 90°C to 120°C are tested under 5 ton for 4 mins and it can be confirmed that hot-press temperature of 110°C showed the highest power reached up to 184 mW. The reason is the temperature plays an important role to provide sufficient and good contact between the catalyst layer and the GOSPEEK membrane at other specific hot-press parameters. Nevertheless, the performance of MEA decreases as the temperature reached 120°C due to the fact that it causes a decrease of porosity and the mass transport rates in the electrodes which increases the diffusion resistance of the GOSPEEK membrane [17].

Figure 6 (a and b) below shows the polarization curves and power curves of hot-pressing time from 4 minutes until 7 minutes. Based on the obtained results, it is realized that the long hot-pressing time showed significant decreases on the performance of the MEA due to the fact that the MEA became more compact [18]. Moreover, a compact MEA will attribute to increase of diffusion resistance and reduces of mass transfer rate. Overall, the suitable hot-pressing time is 4 minutes and it is believed that it provided good contact between the sandwich of electrodes and membrane, as a result giving the highest power of 184 mW [19].

Figure 7 (a and b) displayed the performance of GOSPEEK MEA hot pressed under three hot-pressing pressures from 5 ton until 7 ton. Among the three variables, the hot-press pressure of 5 ton gave the highest power curve and it was recognized as the optimal hot-press pressure. It can be proven that an optimal hot-press pressure able to provide good interface between the catalyst layer and the GOSPEEK membrane and results to increase in performance of MEA in PEMFC [17], [20]. However, an increase of the hot-pressing pressure (7 ton) was not favoured because it is believed that the over-pressing has distorted the structure of the membrane and catalyst layer [17], [21].

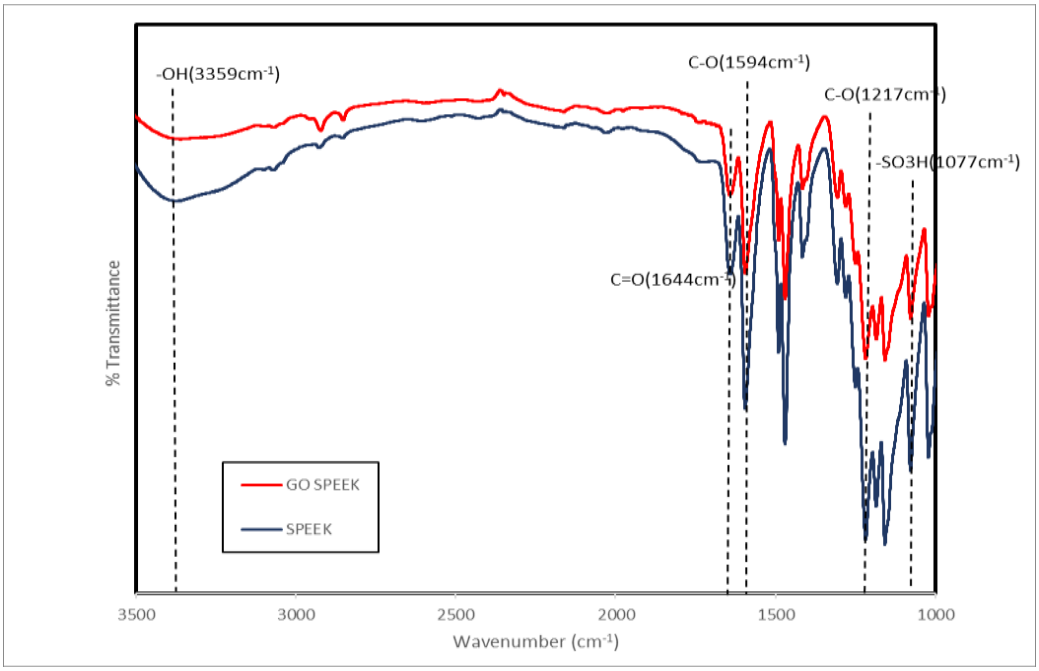


**Figure 3** FT-IR spectra of GOSPEEK and SPEEK

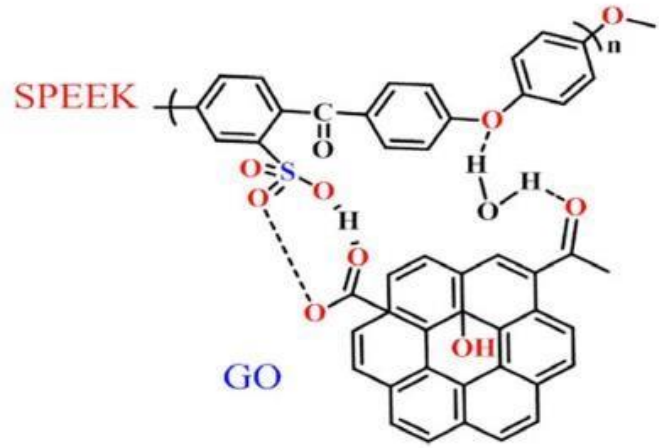


**Figure 4** Schematic diagram of GOSPEEK

### 3.5 Single Fuel Cell Performance

The PEMFC performance of membranes was determined in an external loop single cell under atmospheric temperature. Based on Figure 8, polarization and power curves of GOSPEEK and SPEEK were evaluated to distinguish the performance between the two membranes. Pristine SPEEK membrane indicated the deficit and low power of 22.20 mW which is due to the inconsistency of voltage drop (V) when the current (mA) increases. This could be explained that the pristine SPEEK membrane showing poor chemical stability under the given hot-press parameters, such as 100°C, 5 ton and 4 minutes [22]. Compared to GO SPEEK, the membrane exhibited the power to be 8.30 folds stronger than pristine SPEEK, giving the highest value of 184.02 mW. In this case, the presence of hydrophilic functional groups enhances the proton conductivity and drastically improved the performance of GO SPEEK membrane [22], [23].

This value is comparable to values reported in other studies using similar GO-reinforced membranes [43], [44]. These results confirm that the developed GO-SPEEK membrane performs within a competitive range in terms of electrochemical output.

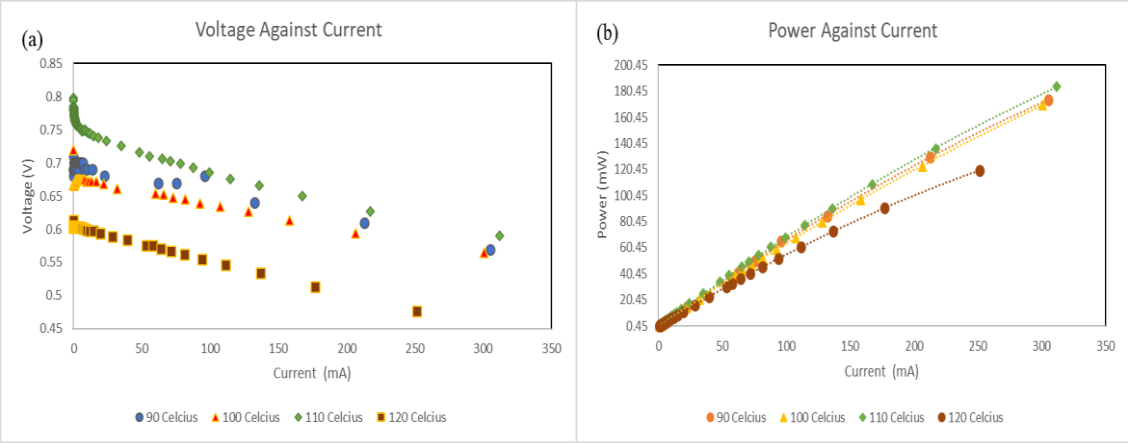


**Figure 5** Scale of hot-pressing temperature on power density and polarization curves under 5 ton for 4 mins

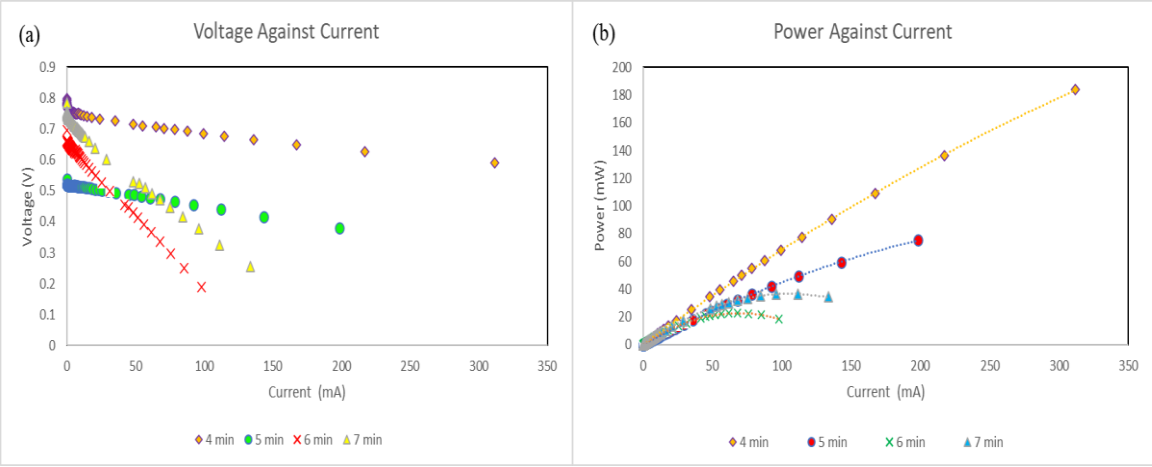


**Figure 6** Scale of hot-pressing time on power density and polarization curves under 5 ton at 110°C

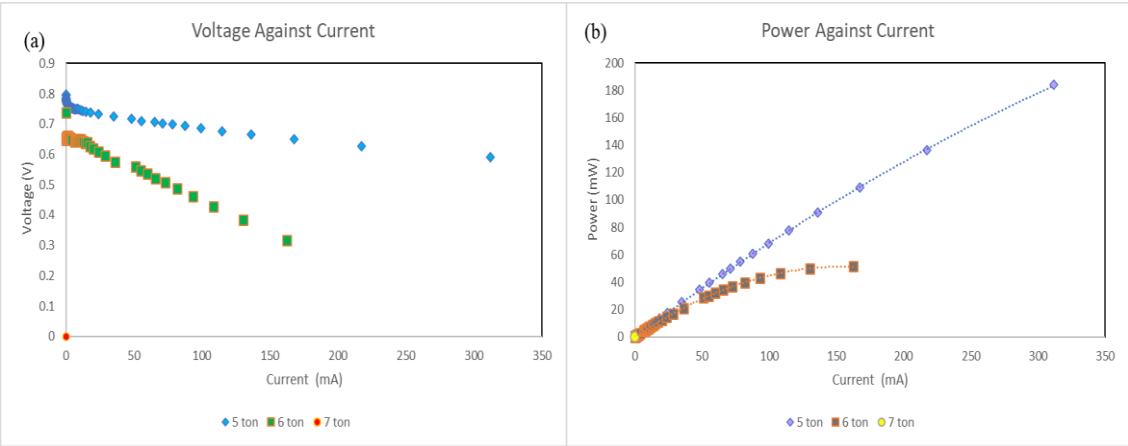


**Figure 7** Scale of hot-pressing pressure on power density and polarization curves at 110°C for 4 min

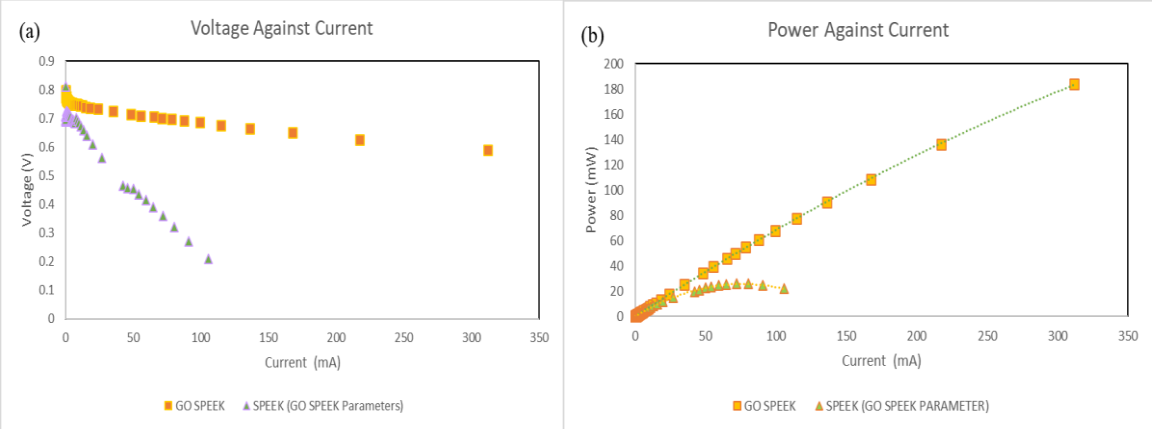


**Figure 8** Polarization curves and power curves of GOSPEEK under optimum hot-press parameters; SPEEK hot-pressed under GOSPEEK optimum parameters

## 4.0 CONCLUSION

In this study, sulfonated polyether ether ketone (SPEEK) and graphene oxide-incorporated nanocomposite membrane (GO-SPEEK) was successfully fabricated for Proton Exchange Membrane Fuel Cells (PEMFCs) application. The addition of graphene oxide (GO) as a carbonaceous filler enhanced the physicochemical properties of the membrane. SEM analysis revealed a rougher, crumpled surface morphology due to the presence of GO, indicating successful integration. The GOSPEEK membrane also demonstrated improved water uptake compared to pristine SPEEK, attributed to the synergistic hydrophilic interaction between the hydroxyl groups of GO and the sulfonic acid groups of SPEEK. FTIR analysis confirmed the presence of these functional groups, which are essential in enhancing proton conductivity and membrane performance. The Membrane Electrode Assembly (MEA) hot-pressed at 110 °C, under 5 tons for 4 minutes, yielded the highest power output of 184.02 mW. Deviations from these optimal hot-pressing conditions led to morphological distortions and performance degradation. Overall, the GOSPEEK membrane fabricated under optimized conditions presents a cost-effective and high-performing alternative to Nafion, with the potential to surpass its efficiency in PEMFC applications.

## ACKNOWLEDGEMENT

The authors express their gratitude to the Ministry of Higher Education (MoHE) for the generous financial support provided under the Long-Term Research Grant Scheme (SATREPS 2023: 1.2) with grant vote number of R.J130000.7809.4L976 (SATREPS 2023: 1.2). This study was also supported by the Science and Technology Research Partnership for Sustainable Development (SATREPS), which is a collaboration effort between the Japan Science and Technology Agency (JST) and the Japan International Cooperation Agency (JICA).

## CONFLICTS OF INTEREST

The author(s) declare(s) that there is no conflict of interest regarding the publication of this paper.

## REFERENCES


[1] Chen, D., Zhou, Z., Liu, H., Gao, Y., Han, D., Wang, Y., & Zhang, J. (2022). Proton exchange membrane fuel cell stack consistency: Evaluation methods, influencing factors, membrane electrode assembly parameters and improvement measures. *Energy Conversion and Management, 261*, 115651. https://doi.org/10.1016/j.enconman.2022.115651.

[2] Sharaf, O. Z., & Orhan, M. F. (2014). An overview of fuel cell technology: Fundamentals and applications. *Renewable and Sustainable Energy Reviews, 32*, 810–853. https://doi.org/10.1016/j.rser.2014.01.012.

[3] Ahmad, S., Nawaz, T., Ali, A., Orhan, M. F., Samreen, A., & Kannan, A. M. (2022). An overview of proton exchange membranes for fuel cells: Materials and manufacturing. *International Journal of Hydrogen Energy, 23*(4), 113–121. https://doi.org/10.1016/j.ijhydene.2022.04.099.

[4] Majlan, E. H., Rohendi, D., Daud, W. R. W., Husaini, T., & Haque, M. A. (2018). Electrode for proton exchange membrane fuel cells: A review. *Renewable and Sustainable Energy Reviews, 89*, 117–134. https://doi.org/10.1016/j.rser.2018.03.007.

[5] Liu, Q., Lan, F., Chen, J., Zeng, C., & Wang, J. (2022). A review of proton exchange membrane fuel cell water management: Membrane electrode assembly. *Journal of Power Sources, 517*, 230723. https://doi.org/10.1016/j.jpowsour.2021.230723.

[6] Xing, P., Robertson, G. P., Guiver, M. D., Mikhailenko, S. D., Wang, K., & Kaliaguine, S. (2004). Synthesis and characterization of sulfonated poly(ether ether ketone) for proton exchange membranes. *Journal of Membrane Science, 229*(1–2), 95–106. https://doi.org/10.1016/j.memsci.2003.09.019.

[7] Elwan, H. A., Mamlouk, M., & Scott, K. (2021). A review of proton exchange membranes based on protic ionic liquid/polymer blends for polymer electrolyte membrane fuel cells. *Journal of Power Sources, 484*, 229197. https://doi.org/10.1016/j.jpowsour.2020.229197.

[8] Wei, P., Zhang, C., Zhang, M., Chen, H., Li, X., Liu, H., & Xie, G. (2022). Sandwich-structure PI/SPEEK/PI proton exchange membrane developed for achieving the high durability on excellent proton conductivity and stability. *Journal of Membrane Science, 644*, 120116. https://doi.org/10.1016/j.memsci.2021.120116.

[9] Suhaimin, N. S., Zulkifli, N. A., Kamarudin, S. K., Masdar, M. S., & Rosli, R. E. (2022). The evolution of oxygen-functional groups of graphene oxide as a function of oxidation degree. *Materials Chemistry and Physics, 278*, 125629. https://doi.org/10.1016/j.matchemphys.2021.125629.

[10] Lin, L. S., Bin-Tay, W., Aslam, Z., Westwood, A. V. K., & Brydson, R. (2017). Determination of the lateral size and thickness of solution-processed graphene flakes. *Journal of Physics: Conference*

*Series, 902*(1), 012026. https://doi.org/10.1088/1742-6596/902/1/012026.

[11] Raduwan, N. F., Shaari, N., Kamarudin, S. K., Masdar, M. S., & Yunus, R. M. (2022). An overview of nanomaterials in fuel cells: Synthesis method and application. *International Journal of Hydrogen Energy.* https://doi.org/10.1016/j.ijhydene.2022.03.035.

[12] Yagizatli, Y., Sahin, A., & Ar, I. (2022). Effect of thermal crosslinking process on membrane structure and PEM fuel cell applications performed with SPEEK-PVA blend membranes. *International Journal of Hydrogen Energy.* https://doi.org/10.1016/j.ijhydene.2022.04.183.

[13] Muniyalakshmi, M., Sethuraman, K., & Silambarasan, D. (2020). Synthesis and characterization of graphene oxide nanosheets. *Materials Today: Proceedings, 21*, 408–410. https://doi.org/10.1016/j.matpr.2019.06.375.

[14] Suhaimin, N. S., Ismail, I., Yusoff, M. M., Harun, Z., Shaari, N., & Othman, N. (2020). Nanocomposite membrane by incorporating graphene oxide in sulfonated polyether ether ketone for direct methanol fuel cell. *Materials Today: Proceedings, 46*, 2084–2091. https://doi.org/10.1016/j.matpr.2021.03.614.

[15] Yin, Y., Wang, H., Cao, L., Li, Z., Li, Z., & Gang, M. (2016). Sulfonated poly(ether ether ketone)-based hybrid membranes containing graphene oxide with acid-base pairs for direct methanol fuel cells. *Electrochimica Acta, 203*, 178–188. https://doi.org/10.1016/j.electacta.2016.04.004.

[16] Therdthianwong, A., Manomayidthikarn, P., & Therdthianwong, S. (2007). Investigation of membrane electrode assembly (MEA) hot-pressing parameters for proton exchange membrane fuel cell. *Energy, 32*(12), 2401–2411. https://doi.org/10.1016/j.energy.2007.07.005.

[17] Yazdanpour, M., Esmaeilifar, A., & Rowshanzamir, S. (2012). Effects of hot pressing conditions on the performance of Nafion membranes coated by ink-jet printing of Pt/MWCNTs electrocatalyst for PEMFCs. *International Journal of Hydrogen Energy, 37*(15), 11290–11298. https://doi.org/10.1016/j.ijhydene.2012.04.139.

[18] Okur, O., Iyigün Karadağ, Ç., Boyaci San, F. G., Okumuş, E., & Behmenyar, G. (2013). Optimization of parameters for hot-pressing manufacture of membrane electrode assembly for PEM (polymer electrolyte membrane fuel cells) fuel cell. *Energy, 57*, 574–580. https://doi.org/10.1016/j.energy.2013.05.001.

[19] Bhosale, A. C., Ghosh, P. C., & Assaud, L. (2020). Preparation methods of membrane electrode assemblies for proton exchange membrane fuel cells and unitized regenerative fuel cells: A review. *Renewable and Sustainable Energy Reviews, 133*, 110286. https://doi.org/10.1016/j.rser.2020.110286.

[20] 039. Zhang, J., Yin, G. P., Wang, Z. B., Lai, Q. Z., & Cai, K. D. (2007). Effects of hot pressing conditions on the performances of MEAs for direct methanol fuel

cells. *Journal of Power Sources, 165*(1), 73–81. https://doi.org/10.1016/j.jpowsour.2006.12.039.

[21] Hasran, U. A., Kamarudin, S. K., Masdar, M. S., Daud, W. R. W., & Yaakob, Z. (2013). Optimization of hot pressing parameters in membrane electrode assembly fabrication by response surface method. *International Journal of Hydrogen Energy, 38*(22), 9484–9493. https://doi.org/10.1016/j.ijhydene.2012.12.054.

[22] Thuyavan, L., Kumar, S. M., Manikandan, V., Ahamed, A. J., & Anantharaj, S. (2021). Polyaniline decorated graphene oxide on sulfonated poly(ether ether ketone) membrane for direct methanol fuel cells application. *Polymers for Advanced Technologies, 32*(9), 3630–3644. https://doi.org/10.1002/pat.5491.

[23] Gokulakrishnan, S. A., Kumar, V., Arthanareeswaran, G., Ismail, A. F., & Jaafar, J. (2022). Thermally stable nanoclay and functionalized graphene oxide integrated SPEEK nanocomposite membranes for direct methanol fuel cell application. *Fuel, 329*, 125407. https://doi.org/10.1016/j.fuel.2022.125407

[24] Ahmad, S., Nawaz, T., Ali, A., Orhan, M. F., Samreen, A., & Kannan, A. M. (2022). An overview of proton exchange membranes for fuel cells: Materials and manufacturing. *International Journal of Hydrogen Energy.* https://doi.org/10.1016/j.ijhydene.2022.04.099.

[25] Rosli, R. E., Husaini, T., Najmi, M. R., Daud, W. R. W., Zainoodin, A. M., Husaini, M., & Rosli, M. I. (2017). A review of high-temperature proton exchange membrane fuel cell (HT-PEMFC) system. *International Journal of Hydrogen Energy, 42*(14), 9293–9314. https://doi.org/10.1016/j.ijhydene.2016.06.211

[26] Adetayo, A., & Runsewe, D. (2019). Synthesis and fabrication of graphene and graphene oxide: A review. *Open Journal of Composite Materials, 9*(2), 207–229. https://doi.org/10.4236/ojcm.2019.92012.

[27] Sun, L. (2019). Structure and synthesis of graphene oxide. *Chinese Journal of Chemical Engineering, 27*(10), 2431–2437. https://doi.org/10.1016/j.cjche.2019.05.003.

[28] Pei, S., Wei, Q., Huang, K., Cheng, H. M., & Ren, W. (2018). Green synthesis of graphene oxide by seconds timescale water electrolytic oxidation. *Nature Communications, 9*(1), Article 145. https://doi.org/10.1038/s41467-017-02479-z.

[29] Aksoy, C., & Anakli, D. (2019). Synthesis of graphene oxide through ultrasonic assisted electrochemical exfoliation. *Open Chemistry, 17*(1), 581–586. https://doi.org/10.1515/chem-2019-0062.

[30] Singh, R. K., Kumar, R., & Singh, D. P. (2016). Graphene oxide: Strategies for synthesis, reduction and frontier applications. *RSC Advances, 6*(69), 64993–65011. https://doi.org/10.1039/C6RA07626B.

[31] Chasanah, U., Trisunaryanti, W., Triyono, Oktaviano, H. S., & Fatmawati, D. A. (2021). The performance of green synthesis of

graphene oxide prepared by modified Hummers method with oxidation time variation. *Rasayan Journal of Chemistry, 14*(3), 2017–2023. https://doi.org/10.31788/RJC.2021.1436042.

[32] Suhaimin, N. S., *et al.* (2024). Novel siliceous mesoporous phosphotungstic acid embedded chitosan/LSMM polymer membrane for direct methanol fuel cells. *Journal of Electroanalytical Chemistry, 968*, 118510. https://doi.org/10.1016/j.jelechem.2024.118510.

[33] Hasran, U. A., *et al.* (2013). Optimization of hot pressing parameters in membrane electrode assembly fabrication by response surface method. *International Journal of Hydrogen Energy, 38*(22), 9484–9493. https://doi.org/10.1016/j.ijhydene.2012.12.054.

[34] Reyes-Rodriguez, J. L., Escorihuela, J., García-Bernabé, A., Giménez, E., Solorza-Feria, O., & Compañ, V. (2017). Proton conducting electrospun sulfonated polyether ether ketone graphene oxide composite membranes. *RSC Advances, 7*(84), 53481–53491. https://doi.org/10.1039/C7RA10484G.

[35] Suhaimin, N. S., *et al.* (2017). Methanol permeability and properties of polymer electrolyte membrane based on graphene oxide-sulfonated (polyether ether) ketone. *Malaysian Journal of Analytical Sciences, 21*(2), 435–444. https://doi.org/10.17576/mjas-2017-2102-19.

[36] Feng, M., Huang, Y., Cheng, Y., Liu, J., & Liu, X. (2020). Rational design of sulfonated poly(ether ether ketone) grafted graphene oxide-based composites for proton exchange membranes with enhanced performance. *Journal of Membrane Science*, *598*, 117676. https://doi.org/10.1016/j.memsci.2019.117676.

[37] Bhattacharyya, S., Mondal, A., Halder, D., Mondal, N. K., Das, B., & Roy, P. (2018). Comparative assessment on the removal of ranitidine and prednisolone present in solution using graphene oxide (GO) nanoplatelets. *Desalination and Water Treatment*, *132*, 333–344. https://doi.org/10.5004/dwt.2018.23170.

[38] He, X., Zhou, Z., Han, Z., Zeng, Y., Chen, X., & Su, J. (2019). Mechanism of controlled release of vancomycin from crumpled graphene oxides. *ACS Omega*, *4*(7), 12439–12447. https://doi.org/10.1021/acsomega.9b00873.

[39] Ramli, W. (2022, April 3). *Water permeability study of Nafion 117 membrane after undergoing hot press*. Academia.edu. Retrieved July 28, 2025, from https://www.academia.edu/75367341/Water_permeability_study_of_nafion_117_membrane_after_undergoing_hot_press.

[40] Krishna, A., Lindhoud, S., & de Vos, W. M. (2021). Hot-pressed polyelectrolyte complexes as novel alkaline stable monovalent-ion selective anion exchange membranes. *Journal of Colloid and Interface Science, 593*, 11–20. https://doi.org/10.1016/j.jcis.2021.02.077.

[41] Vinothkannan, M., Kannan, R., Kim, A. R., Kumar, G. G., Nahm, K. S., & Yoo, D. J. (2016). Facile

enhancement in proton conductivity of sulfonated poly(ether ether ketone) using functionalized graphene oxide—Synthesis, characterization, and application towards proton exchange membrane fuel cells. *Colloid and Polymer Science, 294*(7), 1197–1207. https://doi.org/10.1007/s00396-016-3877-8.

[42] Feng, M., Huang, Y., Cheng, Y., Liu, J., & Liu, X. (2018). Rational design of sulfonated poly(ether ether ketone) grafted graphene oxide-based composites for proton exchange membranes with enhanced performance. *Polymer, 144*, 7–17. https://doi.org/10.1016/j.polymer.2018.04.037.

[43] Kumar, R., Mamlouk, M., & Scott, K. (2014). Sulfonated polyether ether ketone-sulfonated graphene oxide composite membranes for polymer electrolyte fuel cells. *RSC Advances, 4*(2), 617–623. https://doi.org/10.1039/C3RA42390E.

[44] Gokulakrishnan, S. A., Kumar, V., Arthanareeswaran, G., Ismail, A. F., & Jaafar, J. (2022). Thermally stable nanoclay and functionalized graphene oxide integrated SPEEK nanocomposite membranes for direct methanol fuel cell application. *Fuel, 329*, 125407. https://doi.org/10.1016/j.fuel.2022.125407.