

Incorporation of poly(vinylidene fluoride) in sulfonated poly(ether ether ketone) matrix for membrane mechanical stiffness

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Abstract. Development of promising polymer based material for the membrane of the fuel cell was crucial to improve its performance and lifetime. The sulfonated polymer physicochemical properties depend on the degree of sulfonation (DS). High DS of sPEEK exhibits high performance, but poor in mechanical properties due to high water absorption. So, this present study aimed to investigate the effect of incorporating polyvinylidene fluoride (PVDF) at different content toward improvement of high DS sPEEK mechanical properties and ion conduction. The blending approach was used to mix the sPEEK with PVDF (content: 5 wt%; 10 wt%; 15 wt% and 20 wt%). The blended sPEEK/PVDF membrane were prepared by casting and drying the solution using the phase inversion technique. The result showed that the incorporation of PVDF in sPEEK matrix had improved the mechanical properties by reducing water uptake, swelling degree, and dimensional change. Other than that, the tensile strength of blended membranes also higher than neat sPEEK. The proton conductivity of sPEEK/PVDF 15 (7.23 mS/cm) was quite higher and comparable with neat sPEEK (10.31 mS/cm) and Nafion membrane (12 mS/cm). In summary, the incorporation of PVDF in high DS of sPEEK had improved the mechanical properties with promising ionic conduction.

1. Introduction

As of late, the public had increased awareness on the importance of using efficient, clean and eco-friendly energy sources of electricity to reduce the environmental pollution. Development of the fuel cell as a potential energy had attracted researcher to conduct a deep study on this electrochemical device. Generally, the fuel cell can be categorized into several types depend on it employed electrolyte and fed fuel as summarized in Table 1. Fuel cell comprised of several cell components such as a membrane/electrolyte, an anode and a cathode to convert the chemical energy of a fuel gas directly into electrical energy via redox reaction as shown in Figure 1 [1]. The membrane is the heart of the fuel cell that influence the performance where it allows ions to be transmitted between the two sides of the fuel cell. The properties of the membrane highly depend on the used material which are perfluorinated polymer (i.e. Nafion), non-fluorinated polymer (i.e. sulfonated poly(ether ether ketone) [2], sulfonated poly(ether sulfone) [3-5], sulfonated poly(ether imide) [6-8] and natural polymer (i.e. cellulose and chitosan) [9-11]. Although, it had been claimed that the Nafion is the best membrane for fuel cell, but it still has a few limitations in which it is expensive due to complicated manufacturing



process and the cell performance depends too much on water that may degrade at high temperature due to dehydration [12].

Membrane based on non-fluorinated polymer is a popular choice in research works due to its low in cost, ease of synthesizing, and promising physicochemical properties. Neat polymer such as poly(ether ether ketone) (PEEK) does not provide any beneficial influence on fuel cells. Thus, the neat polymer needs to be modified through sulfonating or other techniques to open up and enlarging the dead end channels. Commonly, PEEK is sulfonated by introducing sulfonic acid groups (SO_3H) directly onto the backbone via modification or via polymerization of sulfonated monomers [13]. The properties of sulfonated polymer membrane extremely depend on the degree of sulfonation (DS) where, at high DS ($\geq 80\%$) large attachment of SO_3H resulted to high membrane hydrophilicity, water absorption and performance, but low in mechanical properties [2]. It is important for the membrane to have promising mechanical stability for excellent operation at high current density and temperature as well as the higher diffusion rate of water across the membrane. If the membrane does not have enough mechanical properties the membrane will be facing serious swelling and may lead to splitting between membrane and electrode which in turn will destroy the cell.

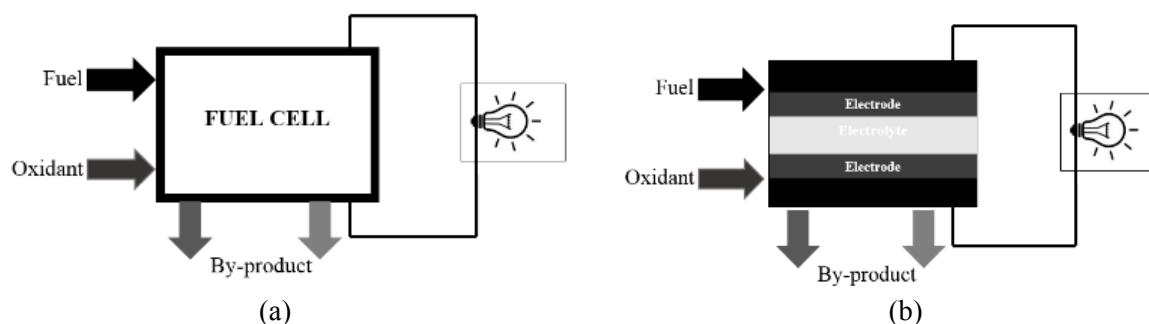


Figure 1. Illustration of fuel cell (a) generates electricity from fuel in one step (b) basic cell components.

Alternatively, the mechanical strength of high DS sPEEK can be improved through two simple approaches which are incorporated with inorganic filler or hydrophobic polymer. The examples of hydrophobic polymer are polyethersulphone (PES), polysulphone (PS), polyetherimide (PEI), poly(bis(phenoxyphosphazene) (POP) and etc. This present study prefers to incorporate PVDF in sPEEK matrix via a blending process as there is only limited previous study only found. Polyvinylidene is an inert/hydrophobic (no charge) polymer that excellence in mechanical, thermal and chemical properties [14]. The present study establishes the blending process between sPEEK and PVDF to form a compatible blend membrane as a potential proton conducting membrane for fuel cell application. The presence of PVDF in sPEEK matrix was varied from 5wt% to 20wt% to determine the promising membrane physicochemical properties based on characterization result.

Table 1. Type of fuel cells [15].

Fuel Cells	Operating temperature	Electrochemical reaction	Electrolyte	Charge carrier in the electrolyte
Alkaline fuel cell (AFC)	60 – 120 °C	Anode: $\text{H}_2 + 2(\text{OH})^- \rightarrow \text{H}_2\text{O} + 2\text{e}^-$ Cathode: $\frac{1}{2} \text{O}_2 + \text{H}_2\text{O} + 2\text{e}^- \rightarrow 2(\text{OH})^-$	KOH solution	$(\text{OH})^-$
Solid alkaline fuel cell (SAFC)	60-90 °C	Anode: $\text{H}_2 + 2(\text{OH})^- \rightarrow \text{H}_2\text{O} + 2\text{e}^-$ Cathode: $\frac{1}{2} \text{O}_2 + \text{H}_2\text{O} + 2\text{e}^- \rightarrow 2(\text{OH})^-$	Solid Polymer	$(\text{OH})^-$
Direct methanol fuel cell (DMFC)	60 – 120 °C	Anode: $\text{CH}_2\text{OH} + \text{H}_2\text{O} \rightarrow \text{CO}_2 + 6\text{H}^+ + 6\text{e}^-$ Cathode: $\frac{3}{2} \text{O}_2 + 6\text{H}^+ + 6\text{e}^- \rightarrow 3\text{H}_2\text{O}$	Solid polymer	H^+
Proton exchange membrane fuel cell (PEMFC)	20 -180 °C	Anode: $\text{H} \rightarrow 2\text{H}^+ + 2\text{e}^-$ Cathode: $\frac{1}{2} \text{O}_2 + 2\text{H}^+ + 2\text{e}^- \rightarrow \text{H}_2\text{O}$	Solid polymer	H^+
Molten carbonate fuel cell (MCFC)	500 - 650 °C	Anode: $\text{H}_2 + \text{CO}_3^{2-} \rightarrow \text{H}_2\text{O} + \text{CO}_2 + 2\text{e}^-$ $\text{CO} + \text{CO}_3^{2-} \rightarrow 2 \text{CO}_2 + 2\text{e}^-$ Cathode: $\frac{1}{2} \text{O}_2 + 2\text{H}^+ + 2\text{e}^- \rightarrow \text{H}_2\text{O}$	Molten carbonate	CO_3^{2-}
Phosphoric acid fuel cell (PAFC)	160 - 200 °C	Anode: $\text{H} \rightarrow 2\text{H}^+ + 2\text{e}^-$ Cathode: $\frac{1}{2} \text{O}_2 + 2\text{H}^+ + 2\text{e}^- \rightarrow \text{H}_2\text{O}$	Phosphoric acid	H^+
Solid oxide fuel cell (SOFC)	600 – 1000 °C	Anode: $\text{H}_2 + \text{O}^{2-} \rightarrow \text{H}_2\text{O} + 2\text{e}^-$ $\text{CO} + \text{O}^{2-} \rightarrow \text{CO}_2 + 2\text{e}^-$ $\text{CH}_4 + 4\text{O}^{2-} \rightarrow 2\text{H}_2\text{O} + \text{CO}_2 + 8\text{e}^-$ Cathode: $\frac{1}{2} \text{O}_2 + \text{CO}_2 + 2\text{e}^- \rightarrow \text{CO}_3^{2-}$	Ceramic compound	O^{2-}

2. Experimental

2.1. Polymer synthesis and membrane preparation methods

The PEEK was sulfonated in concentrated H_2SO_4 at controlled temperature and time that involving the electrophilic substitution reaction [16]. About 50 g of PEEK powder was poured into a beaker containing 1000 ml concentrated H_2SO_4 . The solution was stirred using a mechanical stirrer at 350 rpm for 1 h at room temperature to avoid the agglomeration of polymer. Then, the solution was further stirred for 3 h at 53 °C. This present study used 95-97% concentrated H_2SO_4 instead of 100% H_2SO_4 and chlorosulphuric acid to prevent PEEK polymer degradation and cross-linking reactions [2]. After stirring at the desired time, the sPEEK solution was immediately precipitated in large excess ice-water and filtered. The obtained acidic sPEEK was rinsed several times with deionized water to neutralize at pH 6-7. Lastly, the sPEEK particles were dried in drying oven at 60 °C for 48 h. Figure 2 shows the attachment of sulfonic acid group on PEEK backbone.

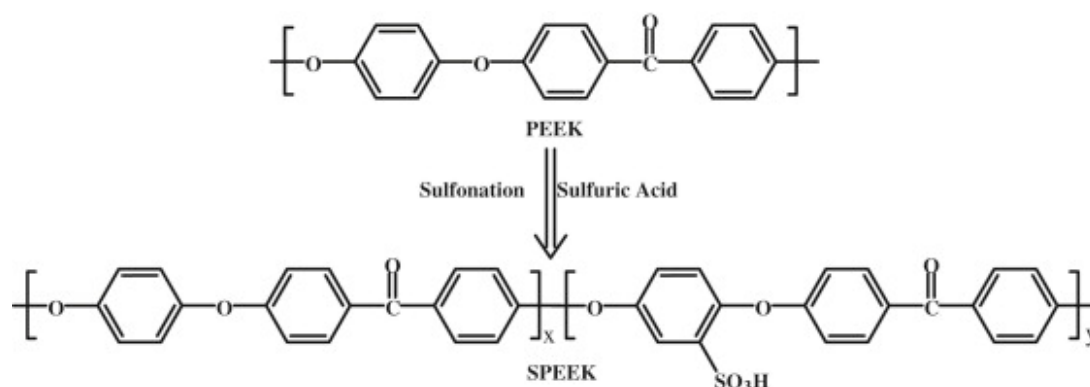


Figure 2. Chemical structure of PEEK and sPEEK [17].

The neat sPEEK membrane was prepared by dissolving 10 g sPEEK powder in 90 ml DMAC to obtain 10 wt% sPEEK solution. It was magnetically stirred at room temperature for 48 h. The completely dissolved sPEEK solution was degassing in a sonicator for 30 min to remove the bubbles and trapped air. Then, the solution was casted on the clean petri dishes and dried in oven at 80 °C for 24 h. The membranes were immersed in water in order to detach it from petri dishes. Then, the sPEEK membranes were dipped in 1 M H₂SO₄ solution for 24 h to activate the sulfonic acid group. Finally, the sPEEK membranes were rinsed several times with water to remove residual acid.

Meanwhile, the sPEEK/PVDF membranes were prepared by magnetically stirred desired amount of sPEEK powder and PVDF into DMAC to produce 10 wt% homogeneous blended solution for 24 h at room temperature. The blended membranes were abbreviated as sPEEK/PVDF (*k*), where *k* represents the volumetric percent of PVDF contents in the blend composition. The composition of sPEEK/PVDF were varied as in Table 2 which are sPEEK/PVDF: 95/5; 90/10; 85/15; and 80/20 in volume. Same procedure as preparation neat sPEEK where the sPEEK/PVDF solution was cast on clean petri dishes and drying in oven for 24 h at 80 °C. The sPEEK/PVDF membranes were peeled off from petri dishes by immersing in water. Then, the membranes were treated with 1 M H₂SO₄ for 24 h at room temperature to activate the sulfonic acid group. Finally, the sPEEK/PVDF membranes were rinsed thoroughly with water to remove residual acid until neutral pH obtained. Figure 3 shows the obtained sPEEK polymer, sPEEK solution, sPEEK/PVDF blended solution and prepared membranes.

Table 2. Concentration of the polymer and solvent.

Membranes	sPEEK (wt%)	PVDF (wt%)	DMAC (wt%)
Neat sPEEK	10.0	0	90.0
sPEEK/PVDF 5	9.5	0.5	90.0
sPEEK/PVDF 10	9.0	1.0	90.0
sPEEK/PVDF 15	8.5	1.5	90.0
sPEEK/PVDF 20	8.0	2.0	90.0

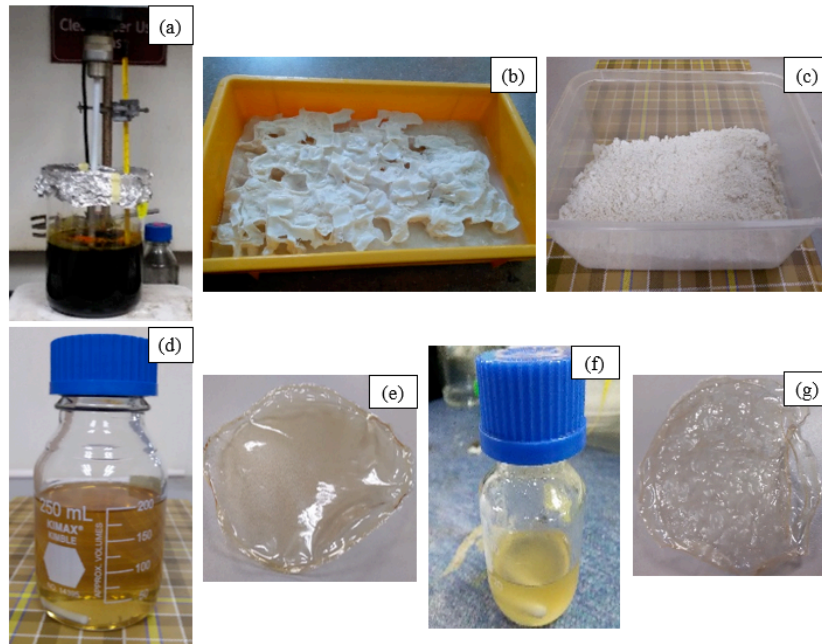


Figure 3. Development of neat sPEEK and sPEEK/PVDF: a) solution of concentrated H₂SO₄ and PEEK, b) precipitated sPEEK in ice-water, c) sPEEK powder, d) solution of DMAC and sPEEK e) neat sPEEK membrane, f) homogeneous solution of sPEEK and PVDF and g) sPEEK/PVDF membrane.

2.2. Characterization Methods

Hydrogen-nuclear magnetic resonance (H-NMR) spectroscopy (Varian Unity Inova, Bruker, Switzerland) modality was used to obtain the DS of synthesis sPEEK powder. The sPEEK powder required to be dried in the oven for 24 h before undergoing H-NMR analysis to trap away the moisture in polymer. The NMR spectrometer was set at 400.13 MHz of resonance frequency at room temperature. The DS of sPEEK was calculated using Equation 1 based on atom numbering in Figure 4 [18].

$$\frac{n}{12-2n} = \frac{AH_A}{\sum AH_{(B,C,D,D',E,E')}} \quad (0 \leq n \leq 1)$$

$$\text{Degree_of_sulfonation (DS)} = n \times 100\% \quad (1)$$

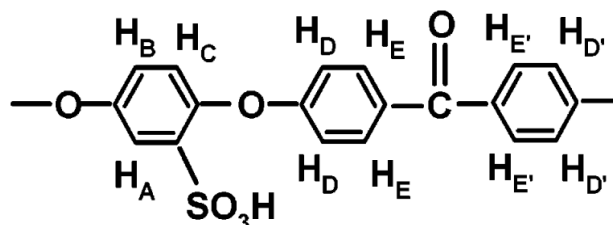


Figure 4. Atom numbering of sPEEK for HNMR analysis.

where n is the number of H_A per repeat unit, the AH_A is the area under the graph of the H_A region and $\sum AH_{(B,C,D,D',E,E')}$ is the total areas under the graph for all of other aromatic hydrogen regions.

Morphological Characterization

The morphological structure of neat sPEEK and sPEEK/PVDF membranes were observed using scanning electron microscope (SEM) (HITACHI TM3030, Japan). SEM is an electron microscope that capture the image of membranes by scanning it surface for determining microstructure image. The membranes were coated with a thin layer of gold via vacuum sputtered before undergoing SEM analysis [18].

Mechanical Properties Characterization

The mechanical properties of developing membrane were determined based on water uptake, swelling ratio, dimensional changes, contact angle, tensile strength, and percentage elongation [19]. The ability of the membranes to absorb the water or water uptake was determined by weighing the weight difference between dried and wet membrane as represent in equation 2. The membranes were dried in a drying oven for 24 h at 60 °C. Then, dried membrane were immersed in deionized water for 24 h at room temperature. The surface of the wet membrane wiped with tissue to remove excess water. The swelling degree of the membrane was determined by measuring the length of dried and wet membranes as shown in equation 3. Meanwhile, the dimensional change was obtained by measuring the thickness of dried and wet membranes (see equation 4).

$$\text{Water uptake} = \frac{m_w - m_d}{m_d} \times 100\% \quad (2)$$

$$\text{Swelling degree} = \frac{S_w - S_d}{S_d} \times 100\% \quad (3)$$

$$\text{Dimensional change} = \frac{D_w - D_d}{D_d} \times 100\% \quad (4)$$

where, m_w is the weight of wet membrane, m_d is the weight of dry membrane, S_w is the length of wet membrane, S_d is the length of dry membrane, D_w is the thickness of the wet membrane and D_d is the thickness of the dried membrane.

The contact angle was observed using a VCA Optima Surface analysis system (AST Products Inc., Billerica, MA) to determine the hydrophobicity and hydrophilicity behavior of the membrane. This analysis is crucial to be conducted because the incorporation of hydrophobic polymer in neat sPEEK may affect the membrane surface wettability. The membranes were resized into 5 cm x 1.5 cm and placed in the holder. Then, a drop of distilled water was pointed at the membrane surface using microsyringe (Hamilton Company, Reno, NV). Before taking the measurements, the distilled water was left in about 30 s periods. Figure 5 shows the illustration of water contact angle measurement.

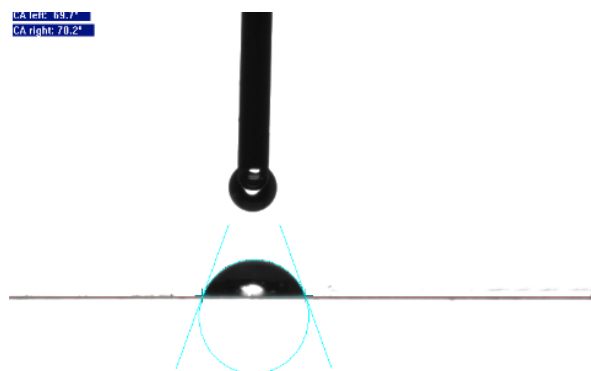


Figure 5. Example of measurement contact angle using VCA optima surface analysis system.

The tensile and percentage elongation of the membrane was measured using mechanical testing system (LRX 5kN, Lloyd Instruments). The membrane was cut into 2.5 cm x 1 cm before testing. Testing system was set at 5 mm min⁻¹ of speed. Equation 6 and 7 represents the tensile strength and percentage elongation.

$$\text{Tensile strength(MPa)} = \frac{\text{Load(N)}}{\text{Gauge width(mm)} \times \text{Membrane thickness(mm)}} \quad (6)$$

$$\text{Percentage elongation} = \frac{\text{Elongation} \times 100}{\text{Gauge length(mm)}} \quad (7)$$

Ionic conduction of membranes

Proton conductivity of the membranes was measured based on two-electrode AC impedance potentiostatic method (AUTOLAB). The frequency was set at range 1 Hz to 10⁵ Hz with 3-50 mV oscillating voltage and tested at room temperature. The membranes were immersed into deionized water more than 24 h to achieve full membrane hydration. The membranes were resized into circular with 18 mm in diameter before placing between two stainless steel block electrodes. Before testing, the thickness of the hydrated membranes was determined using digital micrometer. Proton conductivity, σ of the membranes is represented in the equation 5 [20].

$$\sigma = \frac{T}{RA} \quad (5)$$

where, T represents the thickness of the hydrated membrane, A represents the face area of the membrane and R represents the resistance that determine from the low intersection of the high frequency semi-circle on a complex impedance plane with the Re (Z) axis.

3. Result and Discussion

Degree of sulfonation

According to the HNMR analysis, the sPEEK synthesis in this work was about 80% of DS that considering as having high sulfonic acid group attach to PEEK backbone. Figure 6 shows the spectrum of sPEEK 80%. The sulfonic acid group was successfully attached to PEEK matrix since there is a peak at 7.50-7.55 ppm as reported in Mayahi *et al.*, 2013 [21] study. The percentage attachment of sulfonic acid group can be obtained based on the strength ratio of such a peak to all hydrogen signals where the number of the H_A is exactly equal to the sulfonic acid protons. The sPEEK 80% was soluble in all types of inorganic solvents such as in dimethyl-sulfoxide (DMSO), dimethyl-acetamide (DMAC) and n-methyl-pyrrolidene (NMP).

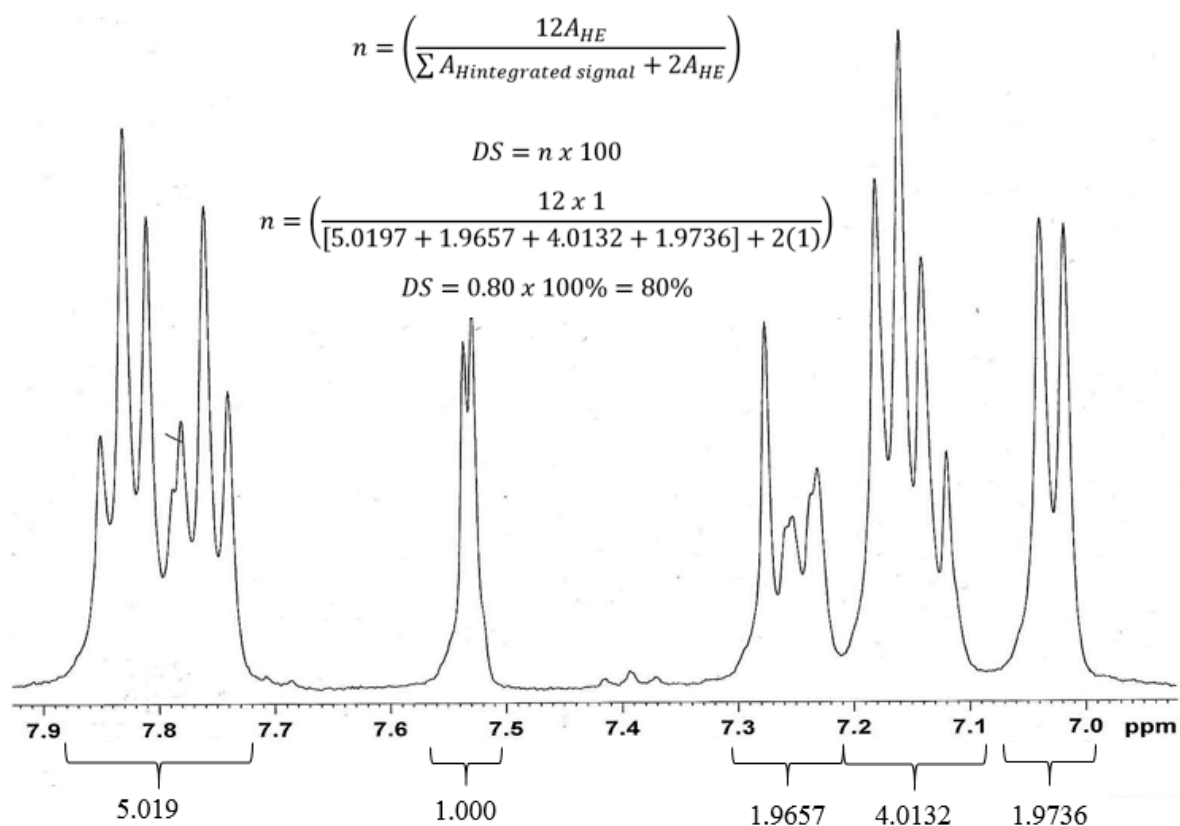


Figure 6. HNMR spectrum of sPEEK.

SEM images

The distribution of PVDF in sPEEK matrix determined from SEM images as well as the morphological structure of neat sPEEK and blend membranes. According to the Figure 7 (a-b), the neat sPEEK membranes were dense, uniform and defect-free, while the PVDF had porous structure. The incorporation of PVDF in sPEEK produce spherically shape in blend membranes as shown in Figure 7 (c-f). Increasing the content of PVDF from 5 to 10 wt% had reduced the formation of sphere in the membrane and it almost disappears as 15 wt% PVDF incorporate in sPEEK. The sPEEK/PVDF 20 membrane had a phase-separated morphology due to incompatibility between those polymers becomes more distinct. Thus, the optimal content of PVDF in sPEEK 80% was 15wt%. The sPEEK/PVDF 15 membrane shown dense, uniform, and finer morphology, which contributed by the homogeneous blending between those polymers. One of the criteria for obtaining good degree compatibility between blending polymer are it had a uniform and a finer morphological structure as observed in sPEEK/PVDF 15. Inan *et al.*, 2010 [22] stated that the PVDF and sPEEK are classified as thermodynamically immiscible because of their dissimilar structures, thus the PVDF should dispersed or a co-continuous morphology to preserve functionality of the membrane. One of the factor that influence the homogeneity of sPEEK/PVDF solution was the variations in the solvent concentration from the surface into the inlet parts [23]. Therefore, the different percentage content of PVDF in sPEEK matrix will produce different number of spherically shape in membrane as shown in morphological structure result. The formation of higher number of spherical shape for sPEEK/PVDF 20 membrane was due to unsuitable amount of PVDF that lead to phase separation. Besides, Inan *et al.*, 2010 [22] stated that the morphology structure influence the ionic domain formation where uniform and finer membrane formed better ionic domains which increase proton conductivity and humidity level.

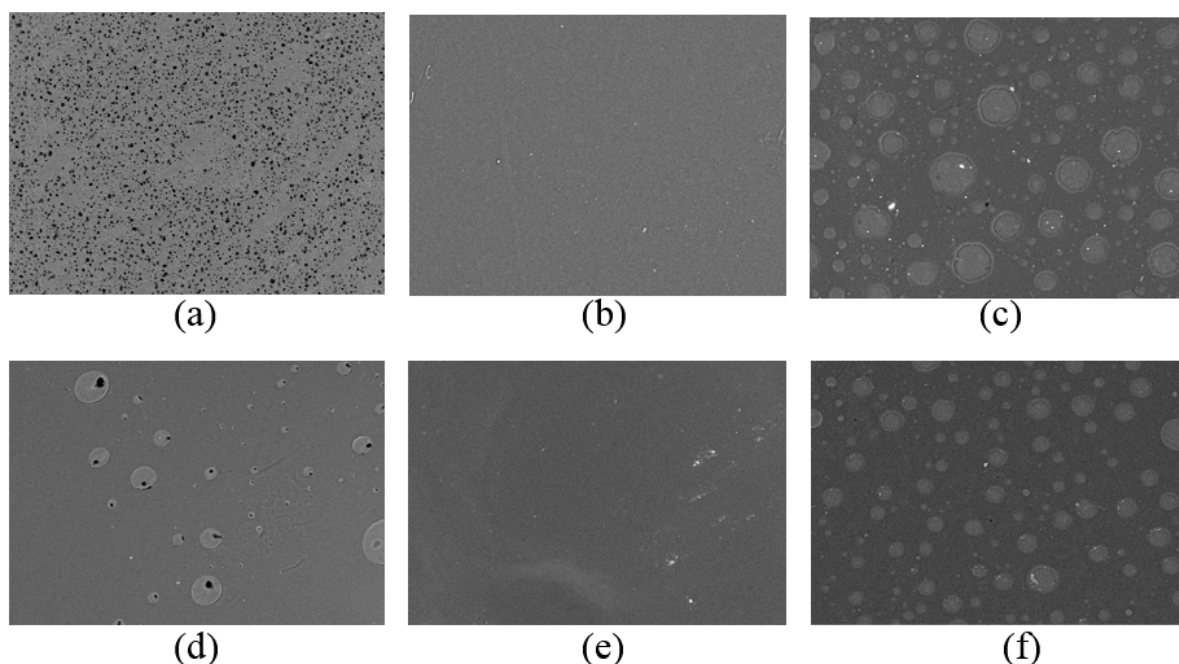


Figure 7. SEM images: a) neat sPEEK, b) PVDF, c) sPEEK/PVDF 5, d) sPEEK/PVDF 10, e) sPEEK/PVDF 15 and f) sPEEK/PVDF 20.

Mechanical properties

Proton conducting membrane required to have a suitable hydration level in order to operate effectively in fuel cells. The optimal hydration levels require to facilitate proton ion transportation from anode side to cathode that known as Grotthuss and vehicular mechanism, besides, to maintain required mechanical properties to be operated in fuel cell especially at high current density and high temperature operation. However, the water absorption by the membrane should be controlled to avoid worse swelling ratio that may cause splitting/loss contact between the membrane and electrodes.

According to Table 3, the highest water uptake, swelling degree and dimensional change was obtained by neat sPEEK 80%. The possible reason was due to the large amount of sulfonic acid group in sPEEK backbone that allow large water absorption. When the PVDF incorporates in sPEEK backbone the percentage of water uptake, swelling degree and dimensional change were reduced due to the hydrophobicity nature of PVDF. As shown in the Table 3, the neat PVDF does not face dimensional change, low water uptake in about 1.96% and small changes in length (5.26%). Thus, the incorporation of PVDF reduces the net hydrophilic nature of the sPEEK 80% blend membranes as found in the contact angle result. Among all of the blend membranes, the sPEEK/PVDF 10 and sPEEK/PVDF 15 absorb more water compared to other although it had more PVDF in sPEEK matrix. This is because it has uniform and finer morphological structure than sPEEK/PVDF 5 and sPEEK/PVDF 20 that facilitate water absorption. In terms of swelling degree, increasing PVDF content had reduced the length changes of the membrane due to increase of hydrophobicity nature that good for it to be operated in fuel cells. The changes in thickness or dimensional change should be higher to provide better contact between membrane and electrode. The neat sPEEK 80% and sPEEK/PVDF 15 exhibits high dimensional change compared to the other membrane which indicate that it had better membrane-electrode contact.

Based on Table 3, it had found the contact angle of neat PVDF was 96.99° that was accepted as hydrophobic nature. The contact angle higher than 90° is consider as hydrophobic material. The neat sPEEK 80% and sPEEK/PVDF membranes had a contact angle in the range of $69.87-77.89^\circ$ which

accepted as hydrophilic nature. The contact angle for this prepared membranes depended on water absorption and morphology structure where the hydrophilicity increase/low contact angle value when water uptake increase and obtain uniform and finer structure. All of the prepared blend membrane having the almost hydrophilic nature with neat sPEEK 80% due to content of PVDF is not too high (less than 50wt%) which does not significantly influence the hydrophilicity behavior of neat sPEEK with high DS.

The tensile strength and percentage elongation of the membranes were determined to investigate its physical mechanical properties when PVDF was incorporated into sPEEK matrix. It is crucial for the proton conducting membrane having excellent physical mechanical properties to increase the lifetime of the fuel cell. Based on the Table 3, the tensile strength and percentage elongation of the membranes increase when the content of PVDF increase. Incorporation of PVDF in sPEEK matrix had improved the structure compactness and intermolecular contact.

Table 3. Mechanical properties of PVDF, neat sPEEK and sPEEK/PVDF membranes.

Types of membrane	Water Uptake (%)	Swelling Degree (%)	Dimensional Change (%)	Contact Angle (°)	Tensile Strength (MPa)	Percentage Elongation (%)
PVDF	1.96	5.26	0.00	96.99	-	-
Neat sPEEK 80%	67.98	16.47	63.27	69.87	21.69	2.51
sPEEK/PVDF 5	57.19	11.11	45.16	74.60	25.24	6.60
sPEEK/PVDF 10	63.80	8.32	40.95	72.25	28.33	6.87
sPEEK/PVDF 15	63.99	6.78	54.01	72.99	32.73	10.33
sPEEK/PVDF 20	46.95	5.78	36.11	77.89	38.52	11.30

Ionic conduction

Figure 8 shows that the highest ionic conduction of blend membranes was found from sPEEK/PVDF 15 (7.23 mS/cm). SPEEK/PVDF membranes obtain low proton conductivity than neat sPEEK but it had better mechanical properties. Incorporation of PVDF had decreased the proton conductivity about 29.87%-60.43% from neat sPEEK. The neat PVDF membrane achieves poor ionic conduction (0.044 mS/cm) because of its hydrophobic nature that only passes less proton ions across the membrane. This might be the reason why the sPEEK membrane with PVDF had a lower proton conductivity than neat sPEEK. However, the proton conductivity of sPEEK/PVDF 15 still considering high and acceptable for fuel cell application and not critical difference with commercial Nafion 117 (percentage difference: 39.75 %). The trend of proton conductivity was related to the water uptake where higher water absorption by membrane lead to high proton conductivity, might be due to involvement of Grotthus and vehicular mechanism that facilitate the ion transport across membranes. Meanwhile, for blend membranes it showed that the morphological structure has influenced the ionic conduction. The dense, finer and uniform structure produce better ionic conduction as is shown by neat sPEEK (10.31 mS/cm) and sPEEK/PVDF 15 (7.23 mS/cm). By considering ionic conductivity as the crucial parameter, this present work chose sPEEK/PVDF 15 as the most promising proton conducting membrane. The 15wt% composition in sPEEK matrix able to produce morphology for the most favorable ion-conduction channel.

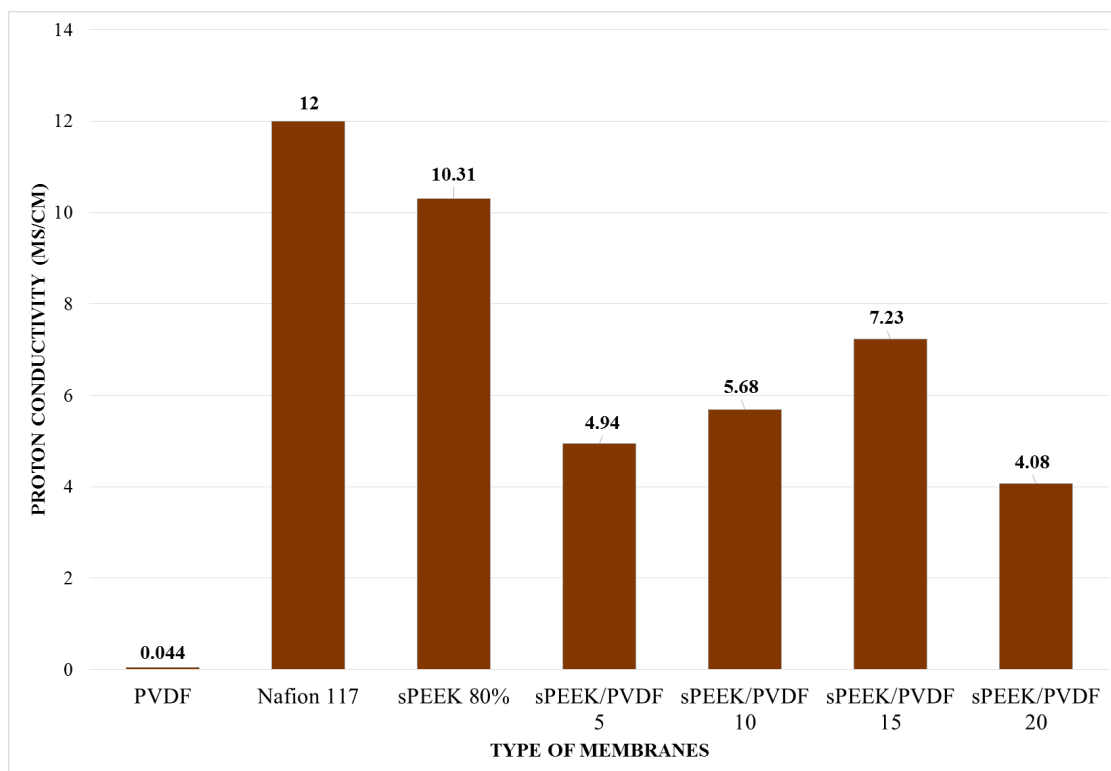


Figure 8. Proton conductivity of neat and blend sPEEK membranes.

4. Conclusion

In this present work, the sPEEK/PVDF membranes were prepared by blending sPEEK 80% with PVDF at 5 wt%, 10 wt%, 15 wt% and 20 wt% content. Sulfonated PEEK with high DS was selected to use in investigating the effect of incorporating PVDF into its mechanical properties and ionic conduction to potentially apply in fuel cell application. This is because the PEEK with high DS ($\geq 80\%$) produce excellent ionic conduction but facing serious swelling degree due to large water absorption that will reduce the cell lifetime. Thus, an alternative should be suggested to overcome its mechanical properties weakness in order to take its advantage in terms of ionic conduction. From this present study, it has been found that the addition of hydrophobic polymer i.e. PVDF is able to improve the mechanical properties of sPEEK 80%. Based on morphological structure and experimental results, the most compatible PVDF content in sPEEK matrix was about 15 wt% due to its uniform and finer structure, besides of excellent mechanical properties and ionic conduction (7.23 mS/cm). Although sPEEK/PVDF 20 had excellent mechanical properties, but it is not suitable to be a proton-conducting membrane for fuel cells due to its poor ionic conduction that is caused by phase separations and non-uniform structure. Generally, the mechanical properties of the membrane were improved when PVDF is incorporated into the sPEEK matrix compared to neat sPEEK. As mentioned earlier, water uptake and proton conductivity depend on the morphological structure of the membranes where their values had increased when the PVDF content increased from 5 wt% to 15 wt%. Nevertheless, the ionic conduction of sPEEK/PVDF is lower than neat sPEEK due to the decrease in hydrophilicity nature, but it is still high and acceptable to be used in fuel cells. In a nutshell, the preparation of a proton-conducting membrane

based on blending process, not only depend on the selection of material but also on the content of hydrophobic polymer incorporate in hydrophilic polymer.

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