



Development of a new highly conductive and thermomechanically stable complex membrane based on sulfonated polyimide/ionic liquid for high temperature anhydrous fuel cells

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ABSTRACT

The paper deals with the synthesis and characterization of a new type of acid doped highly conductive complex membrane based on sulfonated polyimide (sPI) and ionic liquid (IL) for high temperature anhydrous fuel cells. For this purpose, 2,4-diaminobenzene sulfonic acid (2,4-DABSA) is reacted with benzophenontetracarboxylic dianhydride (BTDA) to yield sulfonated poly(amic acid) (sPAA) intermediate. Subsequently, IL is added into sPAA to form an interaction between sulfonic acid and imidazolium group of IL followed by acid doping. The ionic conductivity of acid doped sPI/IL complex polymer membrane is higher than that of IL containing composite membranes reported in the literature ($5.59 \times 10^{-2} \text{ S cm}^{-1}$ at 180°C). Furthermore, dynamic mechanical analysis (DMA) results of acid doped sPI/IL complex membrane show that the mechanical strength of the complex product is slightly changed until 350°C due to the formation of ionic interactions between sulfonic acid groups of sPI and imidazolium groups of IL. Consequently, the ionic interaction not only provides high ionic conductivity with excellent thermomechanical properties (the storage module of 0.91 GPa at 300°C) but also results in a positive effect in long term conductivity stability by blocking IL migration through the membrane.

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1. Introduction

Proton conducting polymer electrolytes have attracted great attentions due to huge demand for sustainable alternative energy sources such as fuel cells (FC) [1]. The perfluorosulfonic acid (PFSA) polymer, Nafion, is one of the most extensively used proton exchange membranes (PEM) for FC applications. However, it has a number of drawbacks such as high cost, high methanol permeability and poor FC performance at temperatures above 80°C due to the loss of water. In FC operations, working at higher temperatures ($100\text{--}200^\circ\text{C}$) provides some advantages such as carbon monoxide (CO) tolerance, easier heat and water management, faster electrode kinetic and high energy efficiency [1–3]. That is why, many research efforts are currently in progress to find new or modified conducting membranes for PEMFC operation between 120 and 150°C . A common approach for improving water retention at higher temperatures in polymer membranes is the incorporating hydrophilic/hygroscopic inorganic additives, such as silica [4,5] and titanium dioxide [6] into membrane matrix. Other additives such as metal phosphates [7,8] (zirconium phosphates, boron phosphate), heteropolyacids [9,10] (phosphotungstic acid, silicotungstic

acid, phosphomolybdic acid, etc.) and phosphoric acid [11] not only increase water uptake but also can provide an extra proton transport pathway by modifying the pore/channel structure of the host polymer. Another approach is the swelling of the membranes with nonaqueous, low-volatile solvents such as imidazole, pyrazole and benzimidazole [12–16]. In addition, Nafion/ionic liquid (IL) composite electrolytes were described and it was found that it is possible to obtain membranes with tailored properties depending on IL cation type [17,18]. Very recently, Choudhury et al. have reported the preparation of a new type of membrane by combining an IL with Nafion reinforced with multifunctional polyhedral oligomeric silsesquioxanes (POSS) using layer-by-layer strategy for high temperature PEMs [19].

Among many advanced polymers, polyimides (PIs) with five and six membered imide rings have been considered as one of the promising candidates for high temperature PEMs because of their excellent thermomechanical properties, chemical stabilities and low methanol permeability [20–27]. In recent years, ILs have attracted extensive attention from different fields such as chemical synthesis, organometallic catalysis, liquid–liquid extractions, analytical techniques and electrochemistry. ILs typically consisting of bulky, asymmetric organic cations and inorganic anions possess many special attractive properties such as non-volatility, non-flammability, high thermal stability, high ionic conductivity and wide electrochemical stability window [28–31].

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In the last few years, polymer electrolytes with IL comprising imidazolium cation have received much attention due to their suitability for HTPEMFC applications [32–43]. Sekhon and Singh have reported the preparation of composite membranes based on polyvinylidene fluoride-hexafluoropropylene (PVDF-HFP) and 2,3-dimethyl-1-octylimidazolium (DMOIm⁺) cation with BF₄⁻, PF₆⁻, CF₃SO₃⁻, N(CF₃SO₂)⁻² anions and they found that the conductivities of the composite polymer electrolytes were in the range of 10⁻³–10⁻⁶ S cm⁻¹ [33]. In another study, Sekhon and lalia have reported the highest conductivity of 0.07 S cm⁻¹ at 120 °C for PVDF-HFP/IL composite membrane with acidic counteranion [34]. In another approach, a series of polymerizable IL was synthesized and polymerized to yield new type of polymer electrolytes by Ohno et al. In that study, polycation type ILs with flexible long spacer showed the ionic conductivity of about 10⁻⁴ S cm⁻¹ at room temperature [37]. On the other hand, there is very scarcely study on the preparation of sulfonated polymer/IL complex membranes and information on their usage in HTPEMFC applications [39–42]. In a fresh article, it was reported that the polymer electrolyte membranes were prepared by the solvent casting method using sPI and 2,3-dimethyl-1-octylimidazolium triflate [dema][TfO] for non-humidified fuel cells and they had high thermal (>300 °C) and proton conductivity (>10⁻² S cm⁻¹ at 120 °C) [41]. However, the long term conductivity stability of the sulfonated polymer/IL complex membranes has not been investigated so far. Only, Scrosati et al. mentioned about the long term operation stability of the polymer based composite membranes with IL and their results showed that IL was largely extracted from the polymer matrix with time [43]. Up to our best knowledge, the long term conductivity stability of the acid doped sPI/IL complex membranes and migration of IL through ionically interacted polymer membranes have not been reported yet.

In this contribution, it is aimed to develop a new highly anhydrous proton conducting and thermomechanically stable complex membrane based on sPI and IL over sulfonated poly(amic acid) (sPAA) intermediate. The main idea with the use of sPI and IL is that the ionic interaction between sulfonic acid groups of sPI and IL's cation could enhance both thermomechanical properties and long term conductivity stability by blocking the IL migration through the polymer matrix. Furthermore, the conductivity and thermomechanical properties of the sPI/IL complex membrane are here reported and discussed in comparison with pristine sPI and literature.

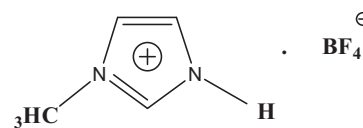
2. Experimental

2.1. Materials

Benzophenonetetracarboxylic dianhydride (BTDA, >97% purity) and 2,4-diaminobenzene sulfonic acid (2,4-DABSA, >97% purity) were used as received from Merck Company, Germany. *N*-Methyl pyrrolidone (NMP), dimethyl acetamide (DMAc), dimethylsulfoxide (DMSO) were supplied from Merck Company, Germany and tetrahydrofuran (THF) was used as received from Carlo Erba, Italy. NMP was purified by distillation under reduced pressure and stored over 5 Å molecular sieve prior using. *N*-Methylimidazol (99%) and tetrafluoroboric acid solution (50% in water) were purchased from Aldrich Company and used without further purification. Sulfuric acid (95–97%) was purchased from Riedel-de Haen Company, Germany.

2.2. Preparation of *N*-methylimidazolium tetrafluoroborate (MeIm-BF₄)

Tetrafluoroboric acid (11.8 ml, 92 mmol, 50% solution in water) was added dropwise to strongly stirred 1-methylimidazole (7.5 g,



N-methyl imidazolium tetrafluoroborate (MeIm-BF₄)

Scheme 1. The chemical structure of MeIm-BF₄.

92 mmol) cooled to 0–2 °C over 30 min. Then, the reaction mixture was stirred for another 2 h and the aqueous solvent was removed in vacuo to yield white waxy product (yield: 98%, elemental analyses results: theoretical; C%: 32.82, H%: 4.42, N%: 16.31, experimental; C%: 32.11, H%: 4.34, N%: 16.18). The chemical structure of MeIm-BF₄ is depicted in Scheme 1.

2.3. Preparation of sPAA

6.44 g of diamine compound (2,4-DABSA) (6 mmol) dissolved in dry NMP/DMSO mixture (1/1 volume) was placed into a 100 ml three-necked round bottomed flask equipped with a thermometer, magnetic stirrer, water bath and nitrogen inlet–outlet system. Subsequently, the equivalent amount of dianhydride compound was slowly added to this solution (20 wt%) at 30 °C and vigorously stirred for 3 h under nitrogen atmosphere to obtain corresponding sulfonated poly(amic acid)(sPAA) intermediate. Then, sPAA solution was kept in a freezer to prevent any further proceeding of the reaction.

2.4. Preparation of sPI/IL complex membrane

IL, namely *N*-methyl imidazolium tetrafluoroborate (MeIm-BF₄), was added into sPAA solution taking sulfonic acid/imidazolium group molar ratio (*n*) of 1.0 and mixed at room temperature for 1 h. Then, ionically interacted sPAA/IL solution was poured into Teflon mold and it was thermally imidized in order to obtain ionically interacted sPI/IL complex membrane. The thermal imidization procedure is given in Scheme 2.

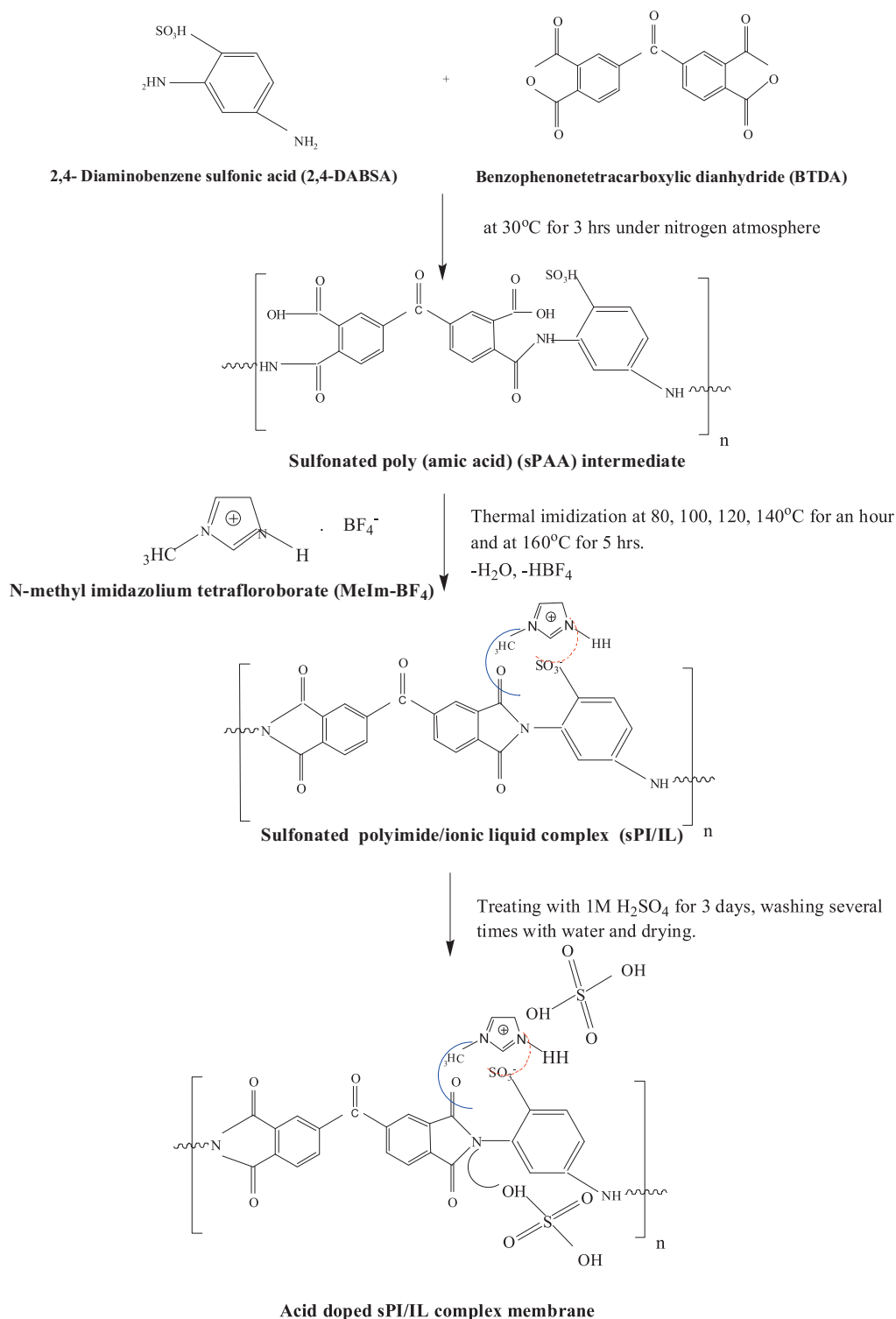
2.5. Preparation of acid doped sPI/IL complex membrane

For the acid doping, sPI/IL complex membrane was treated with 1 M H₂SO₄ for three days. Then, acid doped complex membrane was washed several times with water to remove all excessive acid. After the acid doping, the absorbed amount of dopant was found to be 3 wt%. On the other hand, the samples were stored in room temperature in well sealed glass containers under nitrogen atmosphere to keep them safe from the harmful effect of atmosphere due to the highly hygroscopic nature of IL.

The chemical structures of sPAA, sPI/IL and the proposed ionic interaction between sulfonic acid group of sPAA and imidazolium group of IL (cation) are presented in Scheme 2. The carboxylic acid group of sPAA may also react with IL's cation. However, this interaction will be negligible due to the thermal cyclization of sPAA into sPI structure. As it is shown in Scheme 2, we proposed that the dopant acid can interact with both imide group of sPI and IL cation (MeIm) due to their basicities.

2.6. Characterization

FTIR analyses of acid doped or undoped sPI/IL complex membranes were carried out by Perkin Elmer Spectrum One with ATR (attenuated total reflection) technique. Elemental analyses of IL, namely *N*-methyl imidazolium tetrafluoroborate (MeIm-BF₄), was performed by using Thermo Finnigan Flash EA 1112 Series ele-



Scheme 2. The chemical structures of the sulfonated poly(amic acid) (sPAA), sulfonated polyimide/ionic liquid complex (sPI/IL) and acid doped sPI/IL.

mental analyses equipment. The conductivities of acid doped sPI/IL complex membranes were measured by a two-probe through plane electrochemical impedance spectroscopy using Hewlett Packard-4192A electrochemical workstation over a broad frequency range from 10 kHz to 11 MHz with an amplitude of 5 mV in a wide range of temperature 120–180°C. 10 mm × 10 mm sample was cut and copper wires were contacted by applying silver paste to both sides of acid doped sPI/IL complex membrane with a diameter of 6 mm. Then, the conductivity of acid doped sPI/IL complex membranes

was calculated using the following equation given below.

$$\sigma = \frac{G \times l}{A}$$

where σ is the ionic conductivity (S cm⁻¹), G is the conductance to be determined from admittance plots, l is the thickness of the sample (cm) and A is the cross-section area (cm²) of each electrode. Thermogravimetric analyses (TGA) of acid doped or undoped complex membranes were carried out with a Seiko EXSTAR 6000-

TGA/DTA 6300 model instrument at a heating rate of $10^{\circ}\text{C min}^{-1}$ under nitrogen atmosphere in the temperature range of $40\text{--}800^{\circ}\text{C}$. The samples were dried under vacuum at 60°C prior to measurement for overnight. Dynamic mechanical analysis (DMA) of sPI/IL complex membrane and pristine sPI were performed by using Seiko SII 6100 DMA with $2^{\circ}\text{C min}^{-1}$ heating rate in tension mode. The solubilities of the pristine sPI and acid doped sPI/IL complex membranes were tested by immersing the 0.1 g of membrane into various solvents for 1 week at room temperature.

3. Results and discussion

In this contribution, a new type of highly proton conducting and thermomechanically stable complex membrane based on sPI and IL was developed over sPAA intermediate for the first time as shown in Scheme 2. $-\text{SO}_3\text{H}/\text{imidazolium}$ group molar ratio (n) was chosen as 1.0 for the preparation of ionically interacted sPI/IL complex membrane. Then, the complex membrane was doped with H_2SO_4 to obtain highly conductive sPI/IL complex membranes under non-aqueous conditions. As it is known from the literature, enhanced conductivity can be achieved by doping process using strong acids such as H_2SO_4 and phosphoric acid H_3PO_4 [44]. The idea with the use of sPI and IL is that the ionic interaction between sulfonic acid group and imidazolium groups (IL's cation) not only provides structural reinforcement of the polymer matrix at high temperatures but also limits IL migration which will result in long term conductivity stability.

3.1. FTIR results of sPI/IL complex membrane

Fourier transform infrared (FTIR) spectra of ionically interacted acid doped or undoped sPI/IL complex membranes are shown in Fig. 1. Complete imidization of acid doped sPI/IL complex membrane was confirmed by observation of characteristic imide carbonyl ($\text{C}=\text{O}$) bands at around 1720 cm^{-1} (symmetric imide I) and 1780 cm^{-1} (asymmetric imide I). Also, the absorption band at 1363 cm^{-1} was directly attributed to the characteristic $\text{C}-\text{N}$ stretching of imide groups. The peak observed above 3000 cm^{-1} for acid doped or undoped sPI/IL complex membranes can be attributed to amide groups of imidazolium structure in the complex structure. After the acid doping, the peaks attributed to the protonated amide groups at 1665 and above 3000 cm^{-1} became stronger than that of acid undoped analog. The intensity of $\text{N}-\text{H}$ stretching peak at 3140 cm^{-1} was also increased due to the protonation of both

imide and imidazolium group (MeIm) in the complex structure (see Scheme 2). The absorption bands at 1497 , 1590 , and 1609 cm^{-1} showed the existence of aromatic rings in sPI structure. Furthermore, the absorption bands observed at $1000\text{--}1250\text{ cm}^{-1}$ region, in particular, the strong absorption bands at 1090 and 1196 cm^{-1} , evidenced the $\text{S}-\text{O}$ stretching typical of the sulfonate groups [24]. On the other hand, the peak at around 2930 cm^{-1} was attributed to the presence of aliphatic methyl vibration of MeIm- BF_4 in the complex structure.

3.2. Conductivity of sPI/IL complex membrane

The conductivity of acid doped sPI/IL complex membrane was determined using impedance spectroscopy in the frequency range from 10 kHz to 11 MHz and wide temperature range from 120 to 180°C . Fig. 2 displays the frequency dependence of the alternating current (AC) curves as a function of temperature.

As seen from Fig. 2, the conductivity values of acid doped complex membrane increased with temperature. This enhancement in ionic conductivity was clearly visible at lower frequency region. While the ionic conductivity of acid doped sPI/IL complex membrane was $2.41 \times 10^{-2}\text{ S cm}^{-1}$ at 120°C , this value increased to $5.59 \times 10^{-2}\text{ S cm}^{-1}$ at 180°C (100 kHz). Also, the higher conductivity values were obtained at lower frequencies (10 and 100 kHz) while obtaining slightly lower conductivity values beyond 1 MHz frequency. For an example, the conductivity value of acid doped complex membrane was found to be $2.41 \times 10^{-2}\text{ S cm}^{-1}$ and $2.64 \times 10^{-3}\text{ S cm}^{-1}$ at 10 kHz and 11 MHz , respectively. This slight decrease in the conductivity of acid doped complex membrane with frequency can directly be attributed to the limitation of the dipolar segment movements since polar segments do not orient easily with increasing frequency. The other result can be seen from Fig. 2 that a decrease in conductivity is observed at temperatures higher than 160°C . This result might be due to the evaporation of adsorbed dopant from the complex membrane's surface. Especially, this behavior was clearly observed at higher frequencies due to the limitation of the dipolar segments movements as explained above.

As it is well known, the conductivity of sPI is water dependent and thus the conductivity is very low at high temperature under non-humidified conditions [24]. In the other words, the sulfonic acid group of sPI contributes little to the conductivity or become proton insulator under non-humidified conditions. This result showed us that the conductivity of the complex membrane mainly results from acid doped ionically interacted IL in the membrane. The ionic conductivity of acid doped sPI/IL com-

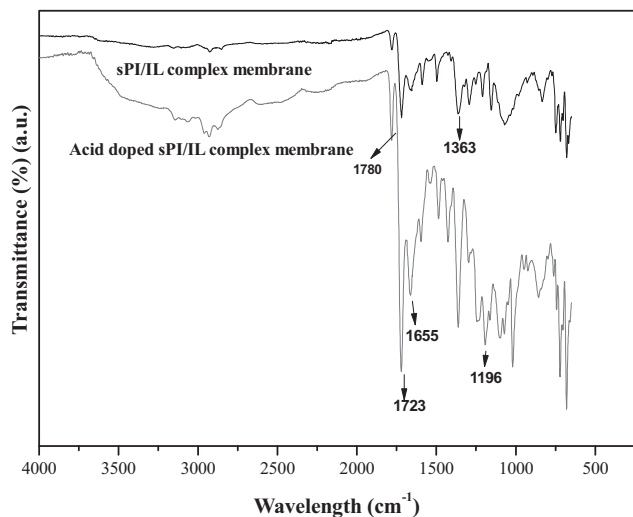


Fig. 1. FT-IR spectra of acid doped and undoped sPI/IL complex membranes.

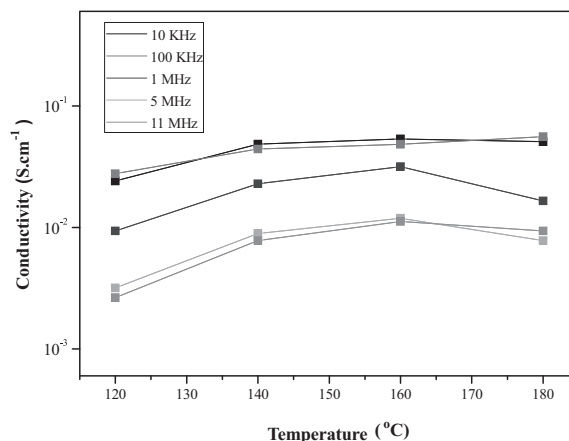


Fig. 2. Conductivity variation of acid doped sPI/IL complex membranes depending on the applied frequency and temperature.

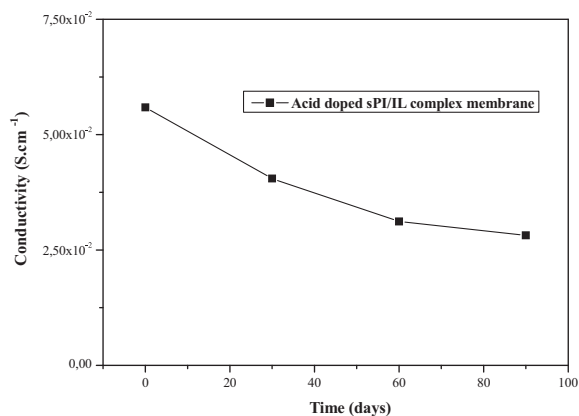


Fig. 3. Time-stability of the ionic conductivity of the acid doped sPI/IL complex membrane (at 180 °C and 100 kHz).

plex membrane was roughly one or two fold higher than that of the composites reported earlier [32,33,36]. In the recent study, Sekhon et al. have found that the ionic conductivity values of IL containing PVdF-co-HFP based composite membranes were in the range of 10⁻³–10⁻⁴ S cm⁻¹ and they emphasized that these results were very promising for anhydrous fuel cell applications [33–36]. According to our conductivity results, it can be clearly concluded that the obtained conductivity values of acid doped sPI/IL complex membrane are higher than that of non-sulfonated polymer matrix system. This enhancement in conductivity can be probably due to (i) the acid doping of both cation of IL (MeIm) and imide group existing in the polymer backbone simultaneously and (ii) the use of diamine compound including sulfonic acid group which can facilitate the ionic conduction through the membrane even if under non-aqueous conditions.

In another set of experiment, the long term conductivity stability of acid doped sPI/IL complex membrane was investigated by repeating the ionic conductivity measurements at 180 °C 90 days later. Fig. 3 presents the conductivity changes of sPI/IL complex membrane with time. Three months later, the conductivity value of sPI/IL complex membrane has dropped from 5.59 × 10⁻² to 2.82 × 10⁻² S cm⁻¹ which is highly sufficient for non-aqueous conditions and promising for high temperature anhydrous FC applications. This decrease in the conductivity of sPI/IL complex membrane may be due to the evaporation of surface adsorbed sulfuric acid. Consequently, it can be clearly emphasized that the ionic interaction between sulfonic acid group of sPI and IL's cation (imidazolium group) provided better long term conductivity stability and blocked IL release by entrapping it into the polymer matrix. In summary, we can conclude that the membranes composed of sPI and IL's cation (MeIm) have the potential to be used as anhydrous electrolytes with long term conductivity stability.

3.3. TGA results of sPI/IL complex membrane

An important criterion of polymer based membranes for high temperature fuel cell applications is their thermal stabilities. For this purpose, TGA analyses of sPI/IL complex membranes were performed in a wide temperature range (30–800 °C) under nitrogen atmosphere. The thermal characteristics of pristine sPI and acid doped sPI/IL complex membrane are presented in Fig. 4 and Table 1 comparatively.

The degradation of acid doped sPI/IL complex membrane took place in 3 steps. The first weight loss was observed at around 100 °C due to the evaporation of adsorbed moisture of the hygroscopic sPIs. The second thermal degradation began at around 210 °C due to the decomposition of both IL's cation and sulfonic acid groups of

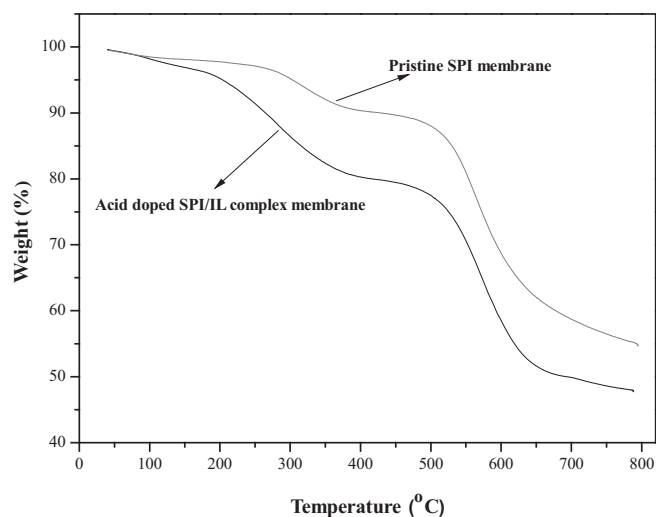


Fig. 4. TGA patterns of pristine sPI and acid doped sPI/IL complex membrane.

Table 1

Thermal degradation values of pristine sPI and acid doped sPI/IL complex membrane.

Membrane	Temperature (°C)		
	T ₅	T ₁₀	T ₅₀
Acid doped sPI/IL complex	203.2	261.9	682.2
Pristine sPI	299.5	404.1	>780.0

T₅: The temperature where 5 wt% weight loss has occurred. T₁₀: The temperature where 10 wt% weight loss has occurred. T₅₀: The temperature where 50 wt% weight loss has occurred.

sPI. Finally, the third degradation step attributed to the decomposition of imide groups in the polymer backbone began above 500 °C. This result was in good agreement with literature [25,35]. Similarly, pristine sPI exhibited three degradation steps. The second one was at around 282 °C, which induced the loss of –SO₃H group while the third one was at around 510 °C corresponded to the decomposition of the polymer main chain. In a recent study, the similar result was reported on IL containing sulfonated poly(ether ether ketone)(sPEEK) based composites by He et al. [42].

3.4. DMA results of sPI/IL complex membrane

The storage (*E'*) and loss (*E''*) modulus of pristine sPI and acid doped ionically interacted sPI/IL complex membrane were analyzed by DMA and the plots are shown in Fig. 5. As seen from the figure, acid doped sPI/IL complex membrane has exhibited slightly

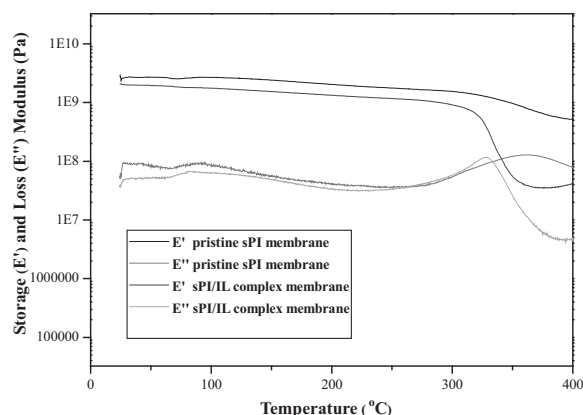


Fig. 5. DMA graphs of pristine sPI and acid doped sPI/IL complex membrane.

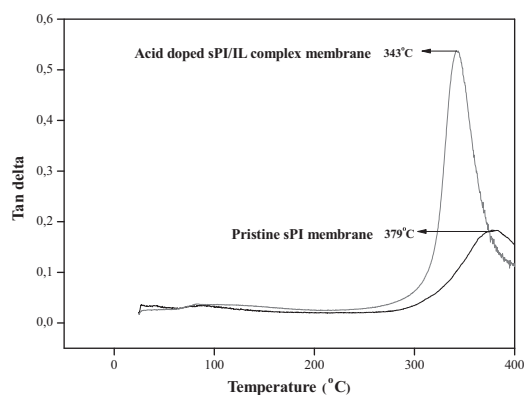


Fig. 6. Tan delta graphs of pristine sPI and acid doped sPI/IL complex membrane.

lower storage and loss modulus than that of pristine sPI. This result may be explained by the presence of IL's cation which presumably decreases the molecular cohesion of the polymer. May be the other reason is that the addition of IL can reduce the inner viscosity of complex electrolytes which makes the segment motion of the polymer chain easier [45]. But, this slight difference in mechanical strength of sPI and sPI/IL complex membrane can be negligible until 300 °C due to the formation of ionic interaction between sulfonic acid group of sPI and imidazolium group of IL. The storage modulus of pristine sPI and acid doped sPI/IL complex membrane were found to be 1.55 GPa and 0.91 GPa at 300 °C, respectively. Recently, Watanabe et al. have reported a new series of polyimide ionomers containing 1*H*-1,2,4-triazole groups in the main chains and sulfonic acid groups in the side chains which they exhibited high maximum stress (>43 MPa at 85 °C and >26 MPa at 120 °C) [41]. Comparing to the literature, it can be concluded that the mechanical strength of sPI/IL complex membrane is higher than that of IL containing sulfonated or non-sulfonated polymer based composite membranes [27,42]. Beyond 320 °C, the storage modulus of acid doped sPI/IL complex membrane rapidly dropped due to the glass transition temperature (T_g) of the complex membrane. T_g can be expressed as the temperature where molecular Brownian motion starts in the polymer structure.

Also, the tan delta curves of these two products are plotted in Fig. 6. The maximum tan delta values for the samples were taken to represent the T_g values. As one can see from the figure that T_g values of pristine sPI and sPI/IL complex membrane were found to be 379 and 343 °C, respectively. The lowering in T_g of the acid doped sPI/IL complex membrane can be directly attributed to the plasticizing effect of IL's cation (MeIm) as explained above.

3.5. Solubility of sPI/IL complex membrane

Solubilities of acid doped complex membrane were tested in different polar solvents such as tetrahydrofurane (THF), *N*-methyl pyrrolidone (NMP), dimethyl sulfoxide (DMSO), dimethyl formamide (DMF), methanol and the results are given in Table 2. These solvents were especially selected in order to investigate the changes in solubility by inserting IL into sPI structure.

Table 2
Solubility of pristine sPI and acid doped sPI/IL complex membrane.^a

Membrane	THF	NMP	DMSO	DMF	MeOH	Ref.
Pristine sPI	–	+	+	+	–	[25]
Acid doped sPI/IL complex	+	+	+	+	–	In this study

THF: tetrahydrofurane, NMP: *N*-methyl-2-pyrrolidone, DMSO: dimethylsulfoxide, DMF: dimethylformamide, MeOH: methanol.

^a Solubility tests were carried out at room temperature.

As seen from Table 2, acid doped ionically interacted sPI/IL complex membrane was completely dissolved in THF, NMP, DMSO and DMF at room temperature. Good solubility properties of the complex membrane may be explained by the presence of IL's cation which may lead a decrease in the molecular cohesion of the polymer. In contrast, both pristine sPI and acid doped sPI/IL complex membrane were not soluble in methanol. This behavior is probably due to the chemical nature of the sulfonated polymer. On the other hand, insolubility of acid doped complex membrane in methanol may offer an alternative material for direct methanol fuel cell (DMFC) applications.

4. Conclusion

In this work, a new type of highly anhydrous conducting complex membrane based on sulfonated polyimide (sPI) and *n*-methyl imidazolium tetrafluoroborate (MeIm-BF₄), ionic liquid (IL), is successfully synthesized over sPAA intermediate for the first time. The complex membrane shows higher ionic conductivity than that of polymer/IL composites reported formerly and a maximum conductivity of $5.59 \times 10^{-2} \text{ S cm}^{-1}$ is achieved at 180 °C. TGA results reveal that new type of acid doped sPI/IL complex membrane can be safely used in high temperature applications up to 200–250 °C. Also, the ionic interaction between both sulfonic acid group of sPI and IL's cation not only provides very high proton conductivity with excellent thermomechanical properties (the storage modulus of 0.91 GPa at 300 °C) but also results in a positive effect in long term conductivity stability by blocking IL migration through the polymer matrix. On the other hand, the high solubility of acid doped sPI/IL complex membrane makes it easily processable.

In summary, we hope that this approach will provide a new way to prepare easily processable sulfonated polymer/IL complex membranes with highly conducting and excellent thermomechanical stability properties over long term for anhydrous PEMFC applications.

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