

Preparation and Characterization of a Semi-Interpenetrating Network Gel Polymer Electrolyte

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ABSTRACT: Poly(PEG200 maleate) was synthesized as a new type crosslinkable prepolymer and the semi-interpenetrating polymer network (semi-IPN) gel electrolytes were prepared by means of thermal polymerization. Their intrinsic properties were characterized by FTIR spectroscopy, differential scanning calorimetry (DSC), X-ray diffractions (XRD), scanning electron microscopy, alternating current impedance (AC impedance), and linear sweep voltammetry. The prepared polymer hosts are transparent and have good mechanical properties. The results of DSC and XRD confirm that the prepared hosts are in amorphous state and

they can hold enough liquid electrolytes, which is favorable for Li⁺ ions to transport via both the absorbed liquid electrolyte and the gel of the entire systems. The semi-IPN gel electrolytes exhibit high ionic conductivity on the order of 10⁻³ S cm⁻¹. Their electrochemical stability up to +4.6 V against Li⁺/Li also makes them potential candidates for application as polymer electrolytes in devices. © 2007 Wiley Periodicals, Inc. *J Appl Polym Sci* 108: 39–46, 2008

Key words: thin films; swelling; cross-linking; electrochemistry

INTRODUCTION

The development of lithium polymer electrolyte batteries has gained an unprecedented significance in the past two decades. Polymer electrolyte is one of the most important components in such system. In 1973, the first measurements on conductivities of poly(ethylene oxide) (PEO) complexes with alkali metal salts were made by Wright and coworkers¹ Since then, the number of contributions to the field of polymer electrolytes has increased enormously.

Until now, there are three types of polymer electrolytes e.g., solid polymer electrolyte (SPE), gel polymer electrolyte (GPE), and composite polymer electrolyte (CPE). Solid solutions of alkali metal salts in polymer (PEO or poly(propylene oxide) (PPO)) are SPE.² The main reason to choose these two polymer hosts is that they can form more stable complexes and possess higher ionic conductivities than any other group of solvating polymers without the addition of organic solvents.³ However, they commonly exhibit conductivities in the range from 10⁻⁸ to 10⁻⁵ S cm⁻¹. The main obstacles to practical applications are, firstly, the high degree of crystallinity, which is unfavorable for ionic conduction in these complexes,

and secondly, the low solubility of salt in the amorphous phases.⁴ CPE is prepared by dispersing of ceramic fillers such as ZrO₂, TiO₂, Al₂O₃, hydrophobic fumed silica, and fiber glass in the polymer matrices.³ The fillers will not only enhance mechanical properties but also lead to higher ionic conductivity because of the enhanced segmental motion of amorphous regions. In addition, the CPE has superior interfacial stability with lithium metal.^{5,6}

GPE is prepared by immobilizing a liquid electrolyte in a polymer matrix and it exhibits high ionic conductivity (10⁻³ to 10⁻⁴ S cm⁻¹).³ The required characteristics of a GPE are as follows: (a) high ionic conductivity (>10⁻³ S cm⁻¹); (b) good mechanical strength; (c) good chemical and electrochemical stability; (d) high thermal stability (up to ~ 90°C); and (e) compatibility with other cell components. The role of the plasticizer in the GPE is to solvate the lithium salt more easily hence to increase the ionic conductivity of the gel electrolytes.⁷ Many kinds of polymer materials, e.g., poly(methyl methacrylate) (PMMA),^{8,9} PEO,¹⁰ poly(acrylonitrile),^{11–16} poly(vinylidene fluoride) (PVdF),^{17–21} poly(vinylidene fluoride-hexa fluoropropylene) (PVdF-HFP)^{22,23} have been carefully studied for GPE. However, all gel polymer hosts will lose their mechanical strength when they are plasticized. Also, the increase of ionic conductivity is always accompanied by the loss of mechanical strength adversely and leads to poor compatibility

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with the lithium electrodes. This high reactivity of lithium metal will result in serious problems in terms of battery cyclability and eventually safety.²⁴

To conquer the drawback of the GPE, researchers tried to prepare by immerse a blending polymers system in the liquid electrolyte. The presence of rigid units of the comparative low polarity polymer will overcome the poor mechanical property of the GPE, while the blending high polarity polymer can absorb abundant liquid electrolyte to maintain high ionic conductivity. Rhoo et al.²⁵ and Stephan et al.^{26–28} have explored to prepare PVC-PMMA blend electrolytes. The problem of poor mechanical strength was circumvented by blending PMMA with polymers such as PVC because of its poor solubility in the plasticizer medium. However, when macromolecules of two distinct types are mixed, large separate phases tend to form with processing conditions in the mixture.

It is well known that full interpenetrating polymer network (IPN) and semi-IPN have been used to improve the compatibility behavior between the immiscible phases.²⁹ In addition, the crosslinked polymer was found to reduce the solubility of the polymer in organic solvents and help to trap liquid electrolyte within the polymer matrix. These polymers can be obtained from monomers that have relatively low molecular weights and are prepared by methods such as ultraviolet radiation, thermal radiation, and electron beam radiation polymerization.³⁰ In this paper, a new type of semi-IPN hosts was prepared by thermal radiation. To get the highly ionic conductive polymer electrolytes with sufficient mechanical strength, the prepared hosts were dipped in an electrolyte solution to activate.

EXPERIMENTAL

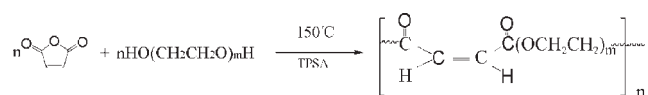
Materials

Poly(ethylene glycol) ($M_n = 200$) (Sinopharm Chemical) and *p*-toluenesulfonic acid (AR) (Sinopharm Chemical) were dried at 80°C under vacuum for 24 h, methyl methacrylate (MMA) (CP)(Sinopharm Chemical) was redistilled under reduced pressure beforehand, THF (AR) (Sinopharm Chemical) and toluene (AR) (Hangzhou Chemical) was dehydrated using 4 Å molecular sieve, maleic anhydride (AR) (Jiangshu Yonghua Chemical). Commercially purchased butadiene-acrylonitrile rubber (NBR) (polybutadiene contents of the NBR, 60%) was plasticated at room temperature before being used.

Preparation of prepolymer

Poly(polyethylene glycol 200 maleate) (PPEG200MA)

It was prepared in the same manner as previously reported.³¹ 0.05 mol maleic anhydride and 0.05 mol



Scheme 1 Synthesis of the poly(polyethylene glycol 200 maleate) (PPEG200MA).

PEG200 were dissolved in toluene, *p*-toluenesulfonic acid (TPSA) used as a catalyst. The mixture was heated to 150°C with magnetic force stirring under argon atmosphere. During the reaction, the outgrowth H_2O was extracted in the water knock-out drum with toluene. The reaction lasted for about 8 h till the amount of calculated H_2O was obtained. Then the product was transferred to a rotary evaporator to remove the solvents. After toluene was evaporated, then the pale yellow gel-like product, poly(polyethylene glycol 200 maleate), was obtained. IR (cm^{-1}): 2877 (ν_{C-H}), 1729 ($\nu_{C=O}$), 1641 ($\nu_{C=C}$), 1401 ($\delta_{as,C-H}$), 1128 ($\delta_{as,C-O-C}$). 1H NMR (in $CDCl_3$) δ (ppm): 6.293 (m, 1H, $-CH=$), 4.343–4.325 (m, 2H, $-CH_2COO-$), 3.742–3.467 (m, 4nH, $-(CH_2CH_2O)_n-$). The reaction involved is outlined in Scheme 1.

PMMA-g-NBR³²

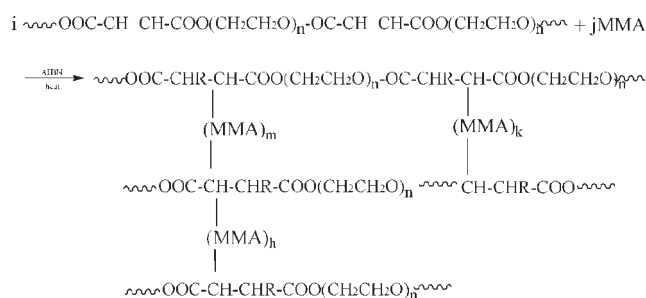
Equal weight of MMA and NBR were dissolved in THF and stirred over night until to become a homogeneous solution, benzoyl peroxide used as an initiator. Grafting reaction was conducted at 75°C under argon shield for 24 h. MMA monomer was assumed to graft onto the NBR chains and form short branch chains under heat. Before completion the reaction, the result solution was transferred to a rotary evaporator to remove solvent; then the resultant was heated up to 110°C to get out the unreacted monomer and the residual THF. To separate the PMMA homopolymer, the prepared grafting copolymer was subsequently extracted with petroleum ether/acetone (1 : 1) for 6 h and dried in a vacuum oven at 60°C for about 8 h. After being dried under reduced pressure at 60°C for 24 h, poly(MMA-g-NBR) was obtained.

IR (cm^{-1}): 2948 (ν_{C-H}), 2237 (ν_{C-N}), 1730 ($\nu_{C=O}$), 1446 ($\delta_{as,C-CN}$), 1149 ($\delta_{as,C-O-C}$) and no absorption peak at 1630 cm^{-1} are observed, indicating no MMA monomer and $C=C$ double bonds in the production. The mol composition of the grafting copolymer is investigated by 1H NMR (in $CDCl_3$) spectrum. δ (ppm): 3.55–3.65 (s, 3H, $-OCH_3$), (2.6–2.65) (m, H, $-CH-CN$). The molar proportion of MMA and AN was deduced to be 1 : 2.96, and the graft degree of PMMA side chain in the system was then calculated to be 25.5 wt % according to the Ref. 33.

Preparation of semi-IPN gel electrolyte

Semi-IPN polymerization system consists of a cross-linkable prepolymer material and uncrosslinked polymer host. In a typical reaction, PMMA-*g*-NBR was first dissolved in THF, and then the viscous solution was cast into a Teflon mold. After evaporating solvent, the PMMA-*g*-NBR films were obtained. PPEG200MA mixed with MMA at a mass ratio of 1 : 4 were used as the crosslinkable prepolymer blends, and azobisisobutyronitrile (1 wt %) was used as an initiator. The mixture was then absorbed in the PMMA-*g*-NBR film. After absorption, the excess prepolymer liquid on the film surface were wiped off with filter paper. The ratio of the absorbed prepolymer blends to PMMA-*g*-NBR was measured by weighting the films before and after soaking, and it was controlled by varying the soaking time of the film in the mixture. Finally, the films were polymerized under N₂ atmosphere at 75°C for 4 h. The cross-linking reaction procedure is shown in Scheme 2. Similarly, the copolymer poly(PPEG200MA-*co*-MMA) was prepared in the same manner, which was heated at 75°C under N₂ atmosphere for 4 h.

The films obtained above were first soaked in methanol for 1 day to remove the unreacted prepolymers, and then dried in a vacuum oven at 60°C for 24 h. To get the semi-IPN gel electrolyte, the dried films were transferred into a glove box and immersed in the organic liquid electrolyte (LB301, DMC : DEC : EC = 1 : 1 : 1 (W/W) LiPF₆ 1M, Guotai-Huarong Chemical) for 0.5 h to activate. After the gel polymer films being removed from the liquid electrolyte solution, excess liquid electrolyte on their surfaces was wiped off with filter paper. The prepared GPE films were flat and homogeneous, and the average GPE films' thicknesses ranged from 70 to 100 μm, which were affected by the preparation process. In this paper, the semi-IPN gel electrolyte with various mass ratio of PMMA-*g*-NBR and poly(MMA-*co*-PPEG200MA) such as 4/1, 3/2, 2/3, and 1/4 were prepared. For convenience, the semi-IPN film and the semi-IPN gel electrolyte obtained with PMMA-*g*-NBR to poly(MMA-*co*-PPEG200MA) mass



Scheme 2 Crosslinking polymerization of PPEG200MA-*co*-MMA.

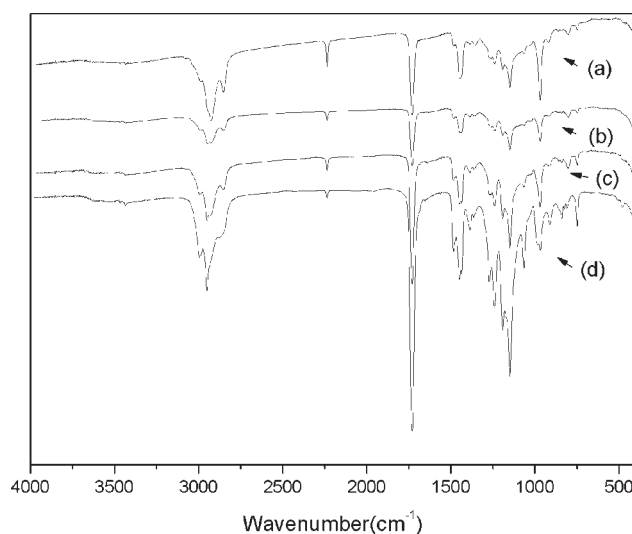


Figure 1 FTIR spectra for semi-IPN films (a) SIPN-N-P-4 : 1; (b) SIPN-N-P-3 : 2; (c) SIPN-N-P-2 : 3; (d) SIPN-N-P-1 : 4.

ratio 4 : 1 were named SIPN-N-P-4 : 1 and G-SIPN-N-P-4 : 1, respectively.

Measurements

FTIR measurements were carried out on BRUKER VECTOR-22 spectrometer at room temperature. The spectra were collected over the range 400–4000 cm⁻¹ by averaging 128 scans at a maximum resolution of 2 cm⁻¹.

Differential scanning calorimetry (DSC) measurements were carried out using a DSC Q100 (TA Instruments, USA) over a temperature of -100°C to 150°C at a scan rate of 10°C min⁻¹. All the thermogram base lines were corrected and calibrated using Indium metal. The experimental specimens (8–10 mg) were dried at 60°C under vacuum for 24 h before being measured. Glass transition temperatures were reported for the midpoint of the transition process. All the samples were firstly annealed at 120°C for 3 min, cooled to -100°C with liquid nitrogen and then scanned for the measurement.

X-ray diffractions (XRD) analysis of the polymer films were performed using a D/MAX-RA X-ray diffractometer (Rigaku) with 2θ values between 3° and 50°.

The cross-sectional morphology of the semi-IPN films were observed by scanning electron microscopy (SEM) (XL30-ESEM, PHILIPS) under vacuum. The prepared films were first dried under vacuum at room temperature for 24 h before being sputtered with gold at 10 mA for 10 min.

Tensile properties of the semi-IPN films were measured through a 10 KN electromechanical tensile testing machine (CMT5104, China) at room temperature. The tensile speed was 40 mm min⁻¹.

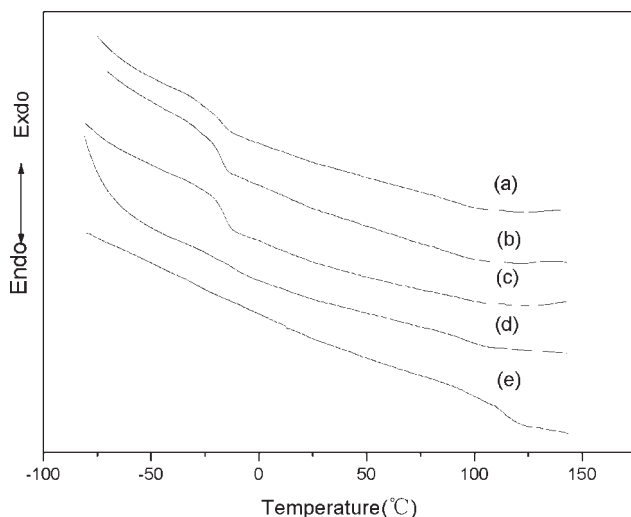


Figure 2 SEM images of semi-IPN films (a) SIPN-N-P-4 : 1; (b) SIPN-N-P-3 : 2; (c) G-SIPN-N-P-2 : 3; (d) SIPN-N-P-1 : 4; and (e) poly(MMA-*co*-PPEG200MA).

Swelling behavior of the semi-IPN films were processed in a dry glove box. The films were first cut into a specimen of 2 cm in diameter. After the mass was weighted, they were immersed in the liquid electrolyte. The semi-IPN films were taken out at different times until no weight changed. The swelling capability of electrolyte solution contents was determined as follows: $S_w = 100 (W - W_0)/W_0$, where W_0 and W were the weights of the initial and wet films.

The ionic conductivity (σ) of the semi-IPN gel electrolytes were determined by AC impedance spectroscopy (EG&G Model 273A potentiostat). It was sandwiched between two parallel stainless steel (SUS) discs (Φ : 1 cm) and mounted in a sealed coin cell to prevent contamination of the sample. The frequency ranged from 100 KHz to 10 Hz at a perturbation voltage of 5 mV. The ionic conductivity (σ) was then calculated from the electrolyte resistance (R_b) obtained from the intercept of the Nyquist plot with the real axis, the film thickness (l), and the electrode area (A) according to the equation $\sigma = l/AR_b$.

The electrochemical stability window of the semi-IPN gel electrolyte was measured by linear sweep voltammetry (LSV) at a scanning rate of 0.05 V s^{-1} . Three-electrode laminated cell was assembled inside a

glove box. Stainless steel (SS) was used as working electrode and lithium metal was used both as a counter and as a reference electrode. LSV measurement was carried out using CHI660A (CH Instruments, Inc).

RESULTS AND DISCUSSION

FTIR spectra of the semi-IPN samples

Figure 1 show the FTIR spectra for the semi-IPN films varied with the mass ratio of poly(PPEG200MA-*co*-MMA) and PMMA-*g*-NBR. The absorption peaks at 1730 and 1100 cm^{-1} are assigned to the contributions from C=O and C—O—C, respectively. Their absorption intensity increases with the crosslinked copolymers. Similarly, the absorption peak at 2236 cm^{-1} is assigned to C \equiv N functional group, which is increased with the content of PMMA-*g*-NBR. The disappearance of the absorption peak for C=C (1640 cm^{-1}) indicates no monomer remained.

DSC measurement

The thermodynamically properties of semi-IPN films and poly(MMA-*co*-PPEG200MA) are characterized by DSC measurement (Fig. 2), and the results are summarized in Table I. The two transitions correspond to the glass transition temperature of the amorphous regions of PMMA-*g*-NBR (T_{g1}) and the crosslinked copolymer (T_{g2}), respectively. T_{g1} is substantially higher than that of pure NBR (-19°C), and it increases with the content of poly(MMA-*co*-PPEG200MA) networks. Reversely, as to T_{g2} , it is decreased with PMMA-*g*-NBR increase. Since T_g is closely related to the flexibility of the polymer chains, and the crosslinking density is an important factor for T_g value. The blending polymer chains of semi-IPN tend to tangle with each other, which will lead to the two T_g values close with each other. The change of T_{g1} and T_{g2} values in Table I illustrates such a trend. In addition, there is no fusion peak presents in the range from -100 to 150°C . This phenomenon contrasts with conventional PEO-based electrolytes, which normally display an endothermic melting peak at around 60°C .³⁴ The absence of a peak is taken as evidence of a fully amorphous

TABLE I
Thermal Transition Temperature for Semi-IPNs

Sample	T_{g1} ($^\circ\text{C}$)	T_{g2} ($^\circ\text{C}$)	ΔH_m (J g^{-1})	χ_c (%)
PMMA- <i>g</i> -NBR/poly(MMA- <i>co</i> -PPEG200MA) = 4 : 1	-17.33	83.36	-	-
PMMA- <i>g</i> -NBR/poly(MMA- <i>co</i> -PPEG200MA) = 3 : 2	-17.13	86.66	-	-
PMMA- <i>g</i> -NBR/poly(MMA- <i>co</i> -PPEG200MA) = 2 : 3	-15.79	90.32	-	-
PMMA- <i>g</i> -NBR/poly(MMA- <i>co</i> -PPEG200MA) = 1 : 4	-11.87	93.98	-	-
Poly(MMA- <i>co</i> -PPEG200MA)	-	114.43	-	-

TABLE II
Mechanical Properties of Semi-IPN Films

Samples	Tensile strength (Mpa)	Fracture strength (Mpa)	Breaking extension (%)
PMMA-g-NBR/poly(MMA-co-PPEG200MA) = 4 : 1	0.53	0.53	371.1
PMMA-g-NBR/poly(MMA-co-PPEG200MA) = 2 : 3	1.84	1.81	153.8
PMMA-g-NBR/poly(MMA-co-PPEG200MA) = 3 : 2	2.23	2.23	96.50
PMMA-g-NBR/poly(MMA-co-PPEG200MA) = 1 : 4	0.77	0.65	4.272

structure in the PEO-like crosslinked polymer network. The maleate unit is thought to act as an irregular pair in PEG chain and hinder the crystallization of PEG. In addition, the crosslinked structure will also prevent the PEO like segments from freely packing into a long-range ordered crystalline structure.³⁵

Stress-strain behavior

Because mechanical strength is an important feature for polymer hosts, their mechanical properties should be addressed.

Table II shows some mechanical properties of the semi-IPN films. For the samples (SIPN-N-P-4 : 1, SIPN-N-P-3 : 2, and SIPN-N-P-2 : 3), there is not any stress concentrations and the compatibility of two blending polymers is very good. The tensile elongation of the samples is enhanced with the content of PMMA-g-NBR. The mechanical properties of these films are the results of the intrinsic nature of the semi-IPN hosts. However, when the weight ratio of the two components is 1 : 4, the breaking extension is only 4.272% and the film is very brittle. It is thought that the excessive crosslinking density will make the blending polymer chains tangle with each other and hard to move freely. The prepared film is very brittle and cannot withstand with the internal pressure build up during practical application. Therefore, the semi-IPN film (SIPN-N-P-1 : 4) will be excluded in further discussions.

XRD measurements

XRD measurements have been carried out for the semi-IPN films and the semi-IPN gel electrolyte in order to examine the nature of the crystallinity for them. Take semi-IPN film (SIPN-N-P-2 : 3) and semi-IPN gel electrolyte (G-SIPN-N-P-2 : 3) as examples.

It can be seen from line (a) and (b) in Figure 3 that there are no sharp crystalline peaks corresponding to PEG in the semi-IPN film and semi-IPN gel electrolyte. The semi-IPN structure can break the orderly arrangement of PEO like segments, and they only show amorphous phases. The result can be interpreted by considering, the Hodge et al.³⁶ criterion which establishes a correlation between the height of the peak and the degree of crystallinity. The results of XRD measurement consist to DSC.

In addition, it is seen from line (b) that no peaks corresponding to pure Li salts of the liquid electrolyte appears in the semi-IPN gel electrolyte which indicates the complete dissolution of liquid electrolyte in the semi-IPN host.

SEM measurements

Typical SEM figures of the surface of the semi-IPN films are present in Figure 4(a-d). Because of PMMA grafted onto NBR chains, the prepolymer blends (PPEG200MA + MMA) can be randomly absorbed within the PMMA-g-NBR films well. And rubber chains can be just trapped in the poly(MMA-co-PPEG200MA) network frame formed by thermal polymerization. Both physical and chemical crosslinking effects make them possible to form rubbery and deformable blends with a fully amorphous structure. The surface of the film shows uniform and homogeneous; hardly any cracks being observed in the figures shows that there is no crystalline structure in the networks. The irregular uneven appearance is thought to be caused by the shrinking effect of the copolymer chains during the polymerization reaction.

Swelling and electrical measurements

Figure 5(a-c) are the typical AC impedance spectra for semi-IPN gel electrolytes varied with the mass

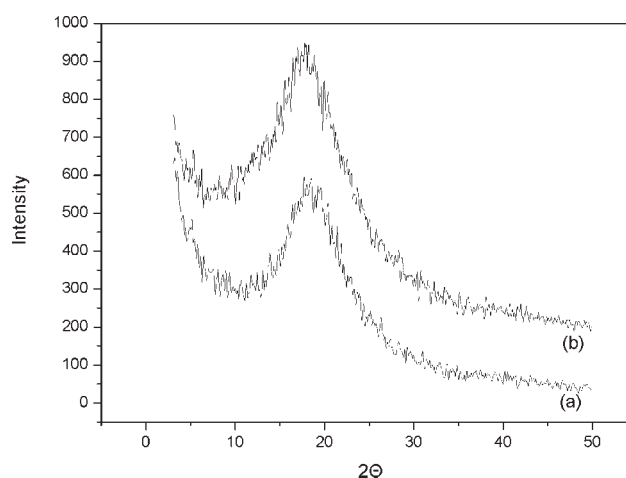


Figure 3 XRD patterns for (a) semi-IPN films (SIPN-N-P-2 : 3); (b) semi-IPN gel electrolyte (G-SIPN-N-P-2 : 3).

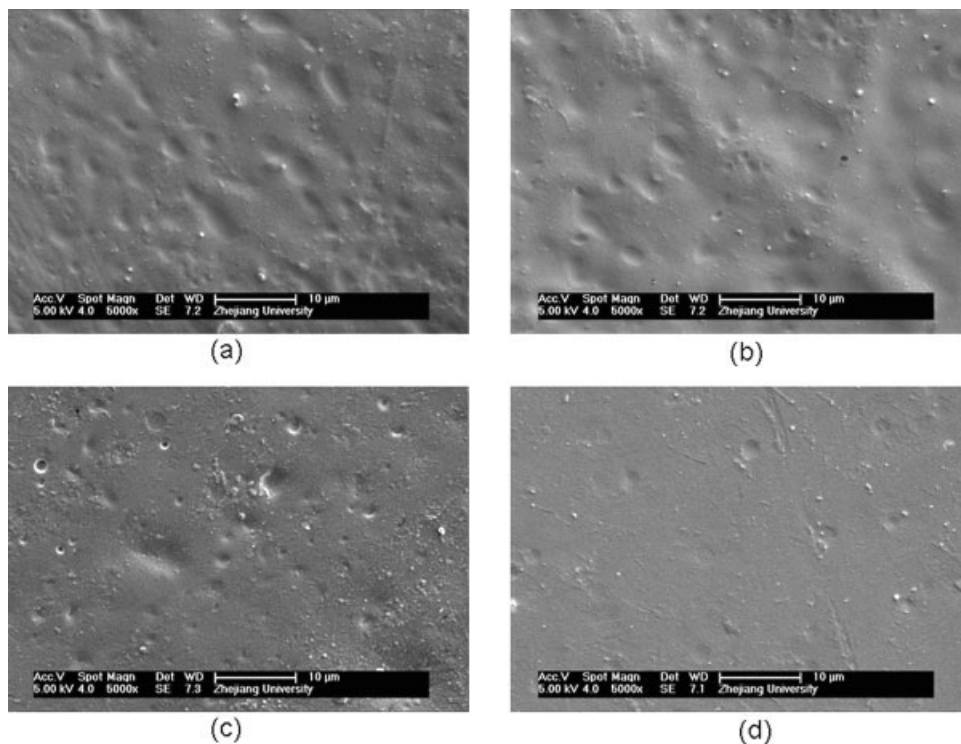


Figure 4 SEM images of semi-IPN films (PMMA-g-NBR/poly(MMA-co-PPEG200MA)): (a) 1 : 4 wt %; (b) 2 : 3 wt %; (c) 3 : 2 wt %; and (d) 4 : 1 wt %.

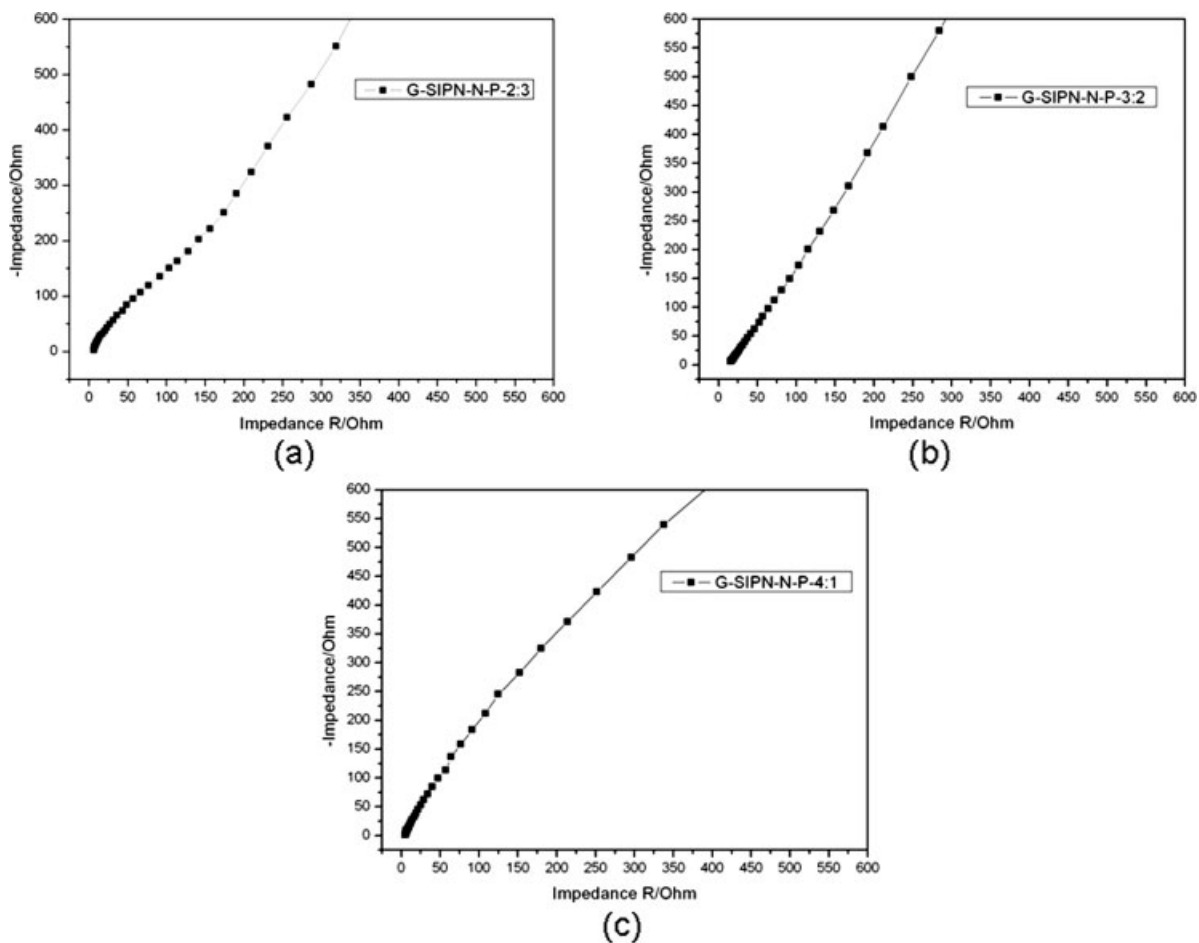


Figure 5 AC impedance spectra of semi-IPN gel electrolytes: (a) G-SIPN-N-P-2 : 3; (b) G-SIPN-N-P-3 : 2; and (c) G-SIPN-N-P-4 : 1.

TABLE III
Properties of Swelling and Ionic Conductivity for Semi-IPN Films

Sample	Area (cm ²)	Thickness (cm)	R_b (Ω)	S_w	σ_T (S cm ⁻¹) ^a
G-SIPN-N-P-4 : 1	0.785	105	6.9	125	0.00194
G-SIPN-N-P-3 : 2	0.785	73	4.14	178	0.00225
G-SIPN-N-P-2 : 3	0.785	80	3.7	222	0.00275

^a At room temperature (30°C).

ratio of two components at 30°C. Behavior of the parallel combination of R_b and C should represent of a characteristic semicircle dispersion of the real (Z') and imaginary ($-Z''$) components of the impedance spectrum. However, a high-frequency semicircle expected for parallel R_b and C is not obtained, it is thought that the semi-IPN gel electrolyte is laid between the two parallel stainless steels and no electrochemical reaction has taken. Only the resistive component of the semi-IPN gel electrolyte could be considered at the high amount of plasticizing electrolyte.

The measured electrochemical properties for the different semi-IPN gel electrolytes at room temperature are given in Table III.

It is seen from Table III that S_w and ionic conductivity at room temperature are dependent on the mass proportion of the two components, which may be due to the materials' polarity and their chemical structure. The methyl methacrylate unit and the crosslinker unit are able to trap a large amount of organic solvent due to good compatibility between the large side functional group $-\text{COOCH}_3$ and carbonate-based organic solvents, such as EC, DMC, and EMC. It is possible to construct an effective local pathway in the absorbed liquid phase and in gel

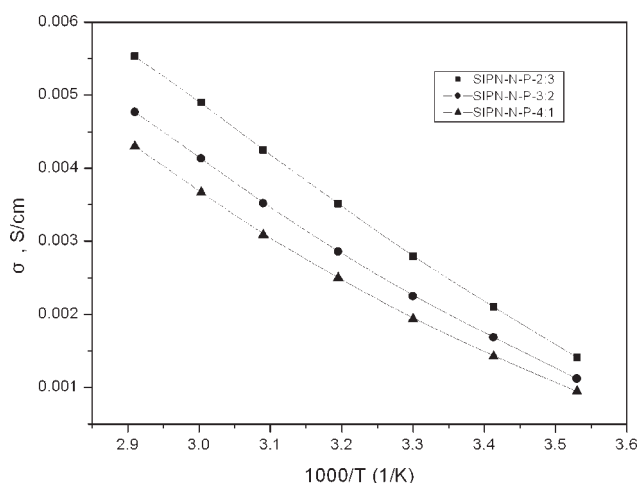


Figure 6 Dependence of conductivity on the reciprocal of temperature for semi-IPN GPE varied with different proportion of blending polymers (wt %).

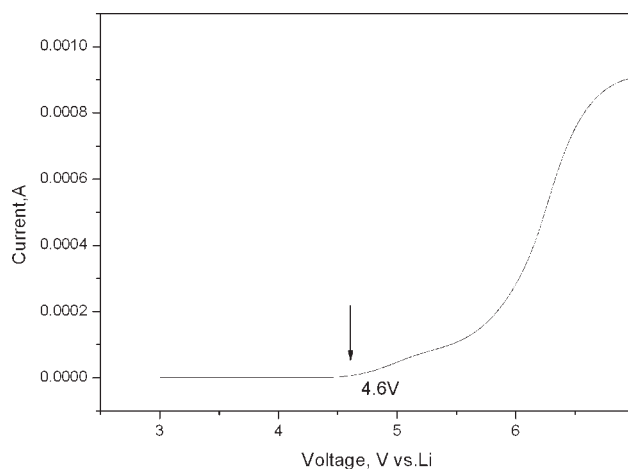


Figure 7 Electrochemical stability window of the semi-IPN gel electrolyte (G-SIPN-N-P-2 : 3) by linear sweep voltammogram.

phase for ionic conduction. PMMA-g-NBR has a comparatively poorer compatibility between organic solvents from thermodynamic considerations. It can produce good mechanical strength in the semi-IPN GPE.

Figure 6 show the ionic conductivity of the semi-IPN gel electrolyte against the reciprocal absolute temperature, a linear relationship confirms that the variation in conductivity with temperature follows an Arrhenius relationship, $\sigma = A \exp(-E/k_B T)$. Where A is a constant which is proportional to the number of carrier ions, k_B represents the Boltzmann constant, and E is the activation energy. The ion mobility in the GPE is decoupled with the segmental motion of the polymer chain, and it is transferred through liquid or gel electrolyte² via an activated hopping mechanism. So it is possible to construct a local effective pathway in the gel phase for ionic conduction. The behavior is typically observed in liquid electrolytes and gel type polymer electrolytes.^{37,38}

Take semi-IPN gel electrolyte (SIPN-N-P-2 : 3) as an example. The LSV of it is presented in Figure 7. The working electrode potential of the cell was varied from 3.0 to 7.0 V (vs. Li) at scanning rate of 0.05 V/s. It is evident from Figure 7 that there is no electrochemical reaction in the potential range from 3.0 to 4.6 V. The onset of current flow at 4.6 V is associated with the decomposition of the electrolyte. From this result, the semi-IPN gel electrolyte is thought to be acceptable for high voltage cathode materials, such as LiCoO_2 , LiNiO_2 , and LiMn_2O_4 .

CONCLUSION

The GPEs with semi-IPN fabric have been synthesized. The prepared polymer hosts present transparent and homogeneous. Relative to the former GPE,

the semi-IPN gel electrolyte cannot only retain the good mechanics, but also absorb a mass of liquid electrolyte to withstand a highly ionic conductivity. Their mechanical and electrochemical properties varied with the content of two components, PMMA-g-NBR and poly(MMA-co-PPEG200MA). When the mass ratio of the two components is 3 : 2, the prepared semi-IPN gel electrolyte shows good properties for application in lithium-ion polymer batteries, and further investigations in application are in progress.

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