

Novel Nanocomposite of Poly(acrylonitrile-co-glycidyl methacrylate) Crosslinked with Jeffamine-Functionalized Multiwalled Carbon Nanotubes as Gel Polymer Electrolytes

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ABSTRACT: Multiwalled carbon nanotubes (MWCNTs) were functionalized with α,ω -diamino poly(propylene oxide) (Jeffamine) of different molecular weights and crosslinked with poly(acrylonitrile-co-glycidyl methacrylate) [P(AN-GMA)] to prepare a novel nanocomposite for applications in gel polymer electrolytes (GPEs). The synthesized copolymer was characterized by $^1\text{H-NMR}$, Fourier transform infrared, and thermal analysis. Scanning electron microscope observation revealed that the Jeffamine-functionalized MWCNTs distributed uniformly in the nanocomposite membrane. The mechanical behaviors of the nanocomposite membranes were investigated. It was found that the crosslinked nanocomposite membranes of P(AN-GMA) and Jeffamine-functionalized MWCNTs exhibited much higher mechanical strength than the counterpart nanocomposite obtained by physical blending. Moreover, the weight content and molecular weights of Jeffamine had an effect on the mechanical properties of the nanocomposites. Differential scanning calorimeter measurements showed that the crosslinked nanocomposite membranes were amorphous. GPEs based on the nanocomposite were prepared and characterized by complex impedance measurements. The GPE based on the nanocomposite of P(AN-GMA) crosslinked with 6 wt % of MWCNTs functionalized by Jeffamine D400 showed an ionic conductivity of about $3.39 \times 10^{-4} \text{ S cm}^{-1}$ at 25°C , which is much higher than the counterpart nanocomposite of physically blended P(AN-GMA) and MWCNTs. © 2012 Wiley Periodicals, Inc. *J. Appl. Polym. Sci.* 000: 000–000, 2012

KEYWORDS: carbon nanotubes; polymer nanocomposites; crosslinking; gel polymer electrolyte

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INTRODUCTION

Gel polymer electrolytes (GPEs) are polymer matrix incorporated with liquid ionic electrolytes, which may be used in rechargeable lithium batteries due to their relatively high ionic conductivity at ambient temperature. In recent years, there have been increasing concerns with safety and energy density of lithium ion batteries, which in turn promoted research interests in the applications of high-performance GPEs in batteries.

Until now, a number of polymers have been investigated as matrix for GPEs, such as poly(ethylene oxide),¹ poly(methyl methacrylate),² polyacrylonitrile (PAN),^{3,4} and poly(vinylidene fluoride).⁵ Of the numerous systems being examined, PAN-based electrolytes have shown interesting characteristics such as high ionic conductivity, thermal stability, and compatibility with the lithium electrodes. It has been proved that the interactions between $-\text{CN}$ groups in PAN and plasticizers as well as with

Li^+ ions in GPEs are responsible for their high ionic conductivity.⁴ However, the addition of plasticizers to these matrixes usually leads to the deterioration in mechanical stability, which may hinder their practical applications in commercial rechargeable lithium ion batteries. Currently, two methods are mainly used to address the problem. Doping the matrix with inorganic materials such as TiO_2 , CeO_2 , MgO , Al_2O_3 , SiO_2 , Zr-O-SO_4 , and molecular sieves ZSM-5^{6–15} by physical blending, is an effective strategy to enhance the mechanical strength and also the ionic conductivity of GPEs. Generally, the used fillers are inert and nonconductive. Chemical crosslinking modification with the formation of covalent bonds is another important means for the improvement of the mechanical properties of GPEs.^{16,17} The resulting crosslinked nanocomposite polymer electrolytes exhibit significantly higher elastic modulus and yield stress than their physically formed nanocomposite counterpart at the cost of negligible loss in transport properties.¹⁸

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Carbon nanotubes (CNTs) are interesting nanomaterial with high surface area and hollow geometry. They have been widely studied because of their remarkable electrical and mechanical properties for various applications including structure-reinforced materials, nanoelectronics, and so forth. The combination of CNTs with polymers is of particular interest as a desirable and facile route to improve the properties of polymers, which is known to largely rely on the homogeneous dispersion of fillers and strong interfacial bonding between the components.¹⁹

Recently, Lee et al. prepared novel electrospun membranes of poly(vinylidene fluoride-*co*-hexafluoropropylene)/multiwalled CNT (MWCNT) composite GPEs. The MWCNTs were modified to improve their dispersion in polymer matrix, and the resulting membranes showed enhanced ionic conductivity at room temperature and good electrochemical and interfacial stability.²⁰

However, GPEs based on the nanocomposite of MWCNTs and polymers in which there are strong chemical bonds between the components have not been reported, despite its potentials in improving the mechanical properties of GPE membrane. In this study, we reported the preparation of GPEs based on the nanocomposite of crosslinkable MWCNTs with poly(acrylonitrile-*co*-glycidyl methacrylate) [P(AN-GMA)]. Specifically, MWCNTs were modified with α,ω -diamino poly(propylene oxide) (PPO) (Jeffamine) of different molecular weights and reacted with P(AN-GMA) to result in a nanocomposite with a crosslinking structure via the formation of covalent bonds. The effect of the chemical crosslinking modification on the morphology, mechanical properties, and ionic conductivity of the resulting GPEs was investigated in detail. A comparison between the nanocomposite with a crosslinking structure and the one prepared by simple physical blending has been attempted.

EXPERIMENTAL

Materials

MWCNTs (diameter: 10–30 nm, length: 5–15 μm , purity >98%) were purchased from Shenzhen Nanotech Port (Shenzhen, China). Thionyl chloride (SOCl_2) (Shanghai Jinshan Tingxin Chemical Reagent, Shanghai, China), sulfuric acid (Zhejiang Juhua, Hangzhou, China), nitric acid (Zhejiang Zhongxing Chemical Reagent, Hangzhou, China) and *N,N*-dimethylformamide (DMF) (Sinopharm Chemical, Shanghai, China) were used as received.

GMA was purchased from Aldrich and used as received. Acrylonitrile (AN (Shanghai Lingfeng Chemical Reagent Company, Shanghai, China) was distilled under reduced pressure. Both GMA and AN were stored in a refrigerator at -20°C . α,ω -Diamino poly(propylene oxide) [Jeffamine D400 ($M = 430 \text{ g mol}^{-1}$) and D2000 ($M = 2000 \text{ g mol}^{-1}$) with the equivalent weights of active hydrogen equal to 115 and 514 g equiv.⁻¹, respectively, Huntsman Corporation, Texas, USA] were dehydrated at 80°C under vacuum for 24 h before use. Lithium electrolyte [(dimethyl carbonate: diethyl carbonate: ethylene carbonate = 1 : 1 : 1 (w/w/w)], LiPF_6 1.0M, (Zhangjiagang Guotai-Huarong New Chemical Materials, Zhangjiagang, China) was stored in a desiccator before use.

Synthesis of MWCNT-Jeffamine

MWCNTs were carboxyl-functionalized as described in reference and denoted as MWCNT-COOH.²¹ The amine-functionalized

MWCNTs were prepared as follows: MWCNT-COOH (1 g) was refluxed in 100 mL of SOCl_2 at 65°C for 24 h, then filtered, washed with tetrahydrofuran (THF), and dried under vacuum at room temperature to give MWCNT-COCl, which was mixed with Jeffamine D400 or Jeffamine D2000 and refluxed at 100°C for 24 h. The resulting amine-functionalized MWCNTs (MWCNT-CONH) were separated by filtration and dried under vacuum, denoted as MWCNT-D400 or MWCNT-D2000 depending on the Jeffamine used.

Synthesis of P(AN-GMA)

Copolymerization of AN and GMA (2 : 1 in weight ratio) was conducted at 70°C using azobisisobutyronitrile as an initiator and DMF as a solvent under N_2 atmosphere for 6 h. On cooling, the mixture was precipitated in methanol, filtered, washed with methanol, and dried *in vacuo* at 40°C for 24 h to obtain the copolymer P(AN-GMA) (denoted as AG for short).

Preparation of the Nanocomposite P(AN-GMA)/MWCNT

The synthetic route of the nanocomposite of P(AN-GMA) and MWCNTs is shown in Scheme 1. P(AN-GMA) was dissolved in DMF with MWCNT-COOH or MWCNT-CONH at different weight ratios and then heated at 80°C for 8 h with vigorous stirring for pre-crosslinking. The resulting mixture was poured onto polytetrafluoroethylene (PTFE) plates, followed by the evaporation of the solvent at room temperature, and heating at 100°C for 24 h to ensure the complete curing. The obtained specimens were denoted as AG/MWCNT-COOH [MWCNT-COOH/P(AN-GMA) = 4 wt %], AG/MWCNT-D400 (4%) [MWCNT-D400/P(AN-GMA) = 4 wt %], AG/MWCNT-D400 (6%) [MWCNT-D400/P(AN-GMA) = 6 wt %], AG/MWCNT-D2000 (4%) [MWCNT-D2000/P(AN-GMA) = 4 wt %], AG/MWCNT-D2000 (6%) [MWCNT-D2000/P(AN-GMA) = 6 wt %], respectively, and dried *in vacuo* at 40°C before use.

Measurements

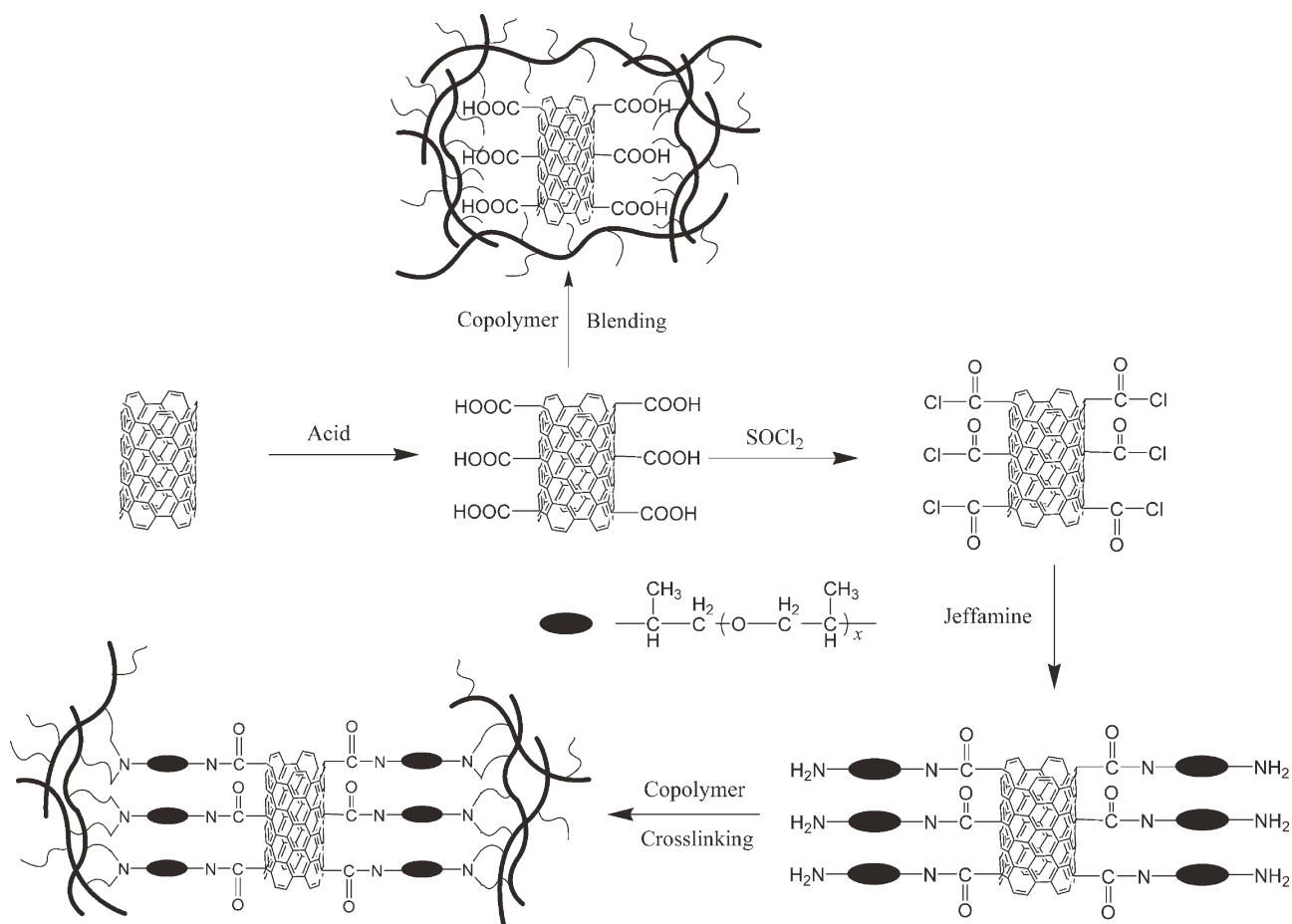
The epoxy equivalent weight (EEW) of P(AN-GMA) was determined by the reaction of a known quantity of the copolymer with a known quantity of hydrochloric acid and back titration of the remaining acid. The EEW was calculated using the following eq. (1):

$$\text{EEW} = \frac{W \times N_v \times 1000}{(B - A) \times F}, \quad (1)$$

where W is the weight of P(AN-GMA) copolymer used, N_v is the sample residue (weight percentage) after heating at 150°C for 30 min, B is the volume of NaOH solution in the blank solution, A is the volume of NaOH solution used in the sample solution, and F is the concentration of NaOH solution.²²

Fourier transform infrared (FTIR) measurements were performed on a BRUKER VECTOR-22 spectrometer (Bruker Corporation, Karlsruhe, Germany). $^1\text{H-NMR}$ spectra were recorded on a Bruker Advance DMX 500 spectrometer (Bruker Corporation, Karlsruhe, Germany) using hexadeuterodimethyl sulfoxide [$(\text{CD}_3)_2\text{SO}$] as the solvent. Morphology of the cross section of the nanocomposite membrane was observed with HITACHI S-4800 scanning electron microscope (SEM).

Thermogravimetric analyses (TGAs) were conducted on a Perkin-Elmer Pyris7 instrument (Perkin-Elmer, Massachusetts, USA) at



Scheme 1. Synthetic process of the nanocomposite of P(AN-GMA) and functionalized MWCNTs.

a heating rate of $20^{\circ}\text{C min}^{-1}$ under nitrogen. Thermal analyses were performed using a TA DSC Q100 differential scanning calorimeter (DSC) (TA Instruments, Delaware, USA) under N_2 atmosphere with a heating rate $10^{\circ}\text{C}\cdot\text{min}^{-1}$ from -50 to 150°C .

The mechanical properties of the nanocomposite membranes were evaluated from the stress-strain tests using an Instron model 5500 testing device (Instron, Massachusetts, USA). Tensile tests were performed under a 1000N load cell at a strain rate of 5 mm min^{-1} .

The ionic conductivity (σ) of the corresponding GPEs was determined by AC impedance spectroscopy (EG&G Model 273A potentiostat, Princeton Instruments, New Jersey, USA). The membrane was sandwiched between two parallel stainless steel discs ($d = 1\text{ cm}$). During the measurement, it was 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 10 mV . The ionic conductivity was calculated from the electrolyte resistance (R_b) obtained from the intercept of the Nyquist plot with the real axis, the membrane thickness (l), and the electrode area (A) according to the equation $\sigma = l/AR_b$.

RESULTS AND DISCUSSION

To obtain chemically crosslinked polymer/MWCNT nanocomposite for GPEs, carboxyl-functionalized MWCNTs were reacted

with SOCl_2 to obtain active carbonyl chloride group, which reacted further with α,ω -diamino PPO to introduce amino groups. Their subsequent reaction with epoxide groups in P(AN-GMA) led to the formation of a crosslinked nanocomposite as illustrated in Scheme 1. As a result, chemical bonding modification was realized between the two components in the nanocomposite. Their structures and morphologies were characterized by different methods as outlined below.

TGA Measurements

Weight loss curves of pristine MWCNTs, MWCNT-D400, and MWCNT-D2000 are shown in Figure 1 for comparison. It is observed that the weight loss curve of pristine MWCNTs below 800°C exhibits a flat slope with a weight loss of $\sim 6.7\%$ between 200 and 450°C . In contrast, there is an obvious weight loss stage between 200 and 450°C for MWCNT-D400 (26.1%) and MWCNT-D2000 (36.8%), which can be mainly attributed to the functional groups-Jeffamine D400 and -Jeffamine D2000 grafted onto the MWCNTs, respectively. Moreover, the weight loss of MWCNTs was also taken into account while calculating the graft ratio of Jeffamines. The grafting efficiency a of Jeffamine onto MWCNTs can be calculated by the formula $a = 12x/M(1-x)$ (where M is the molecular weight of Jeffamine and x is the net weight loss of Jeffamine in TGA by considering the slight weight loss due to MWCNTs). It can be estimated that

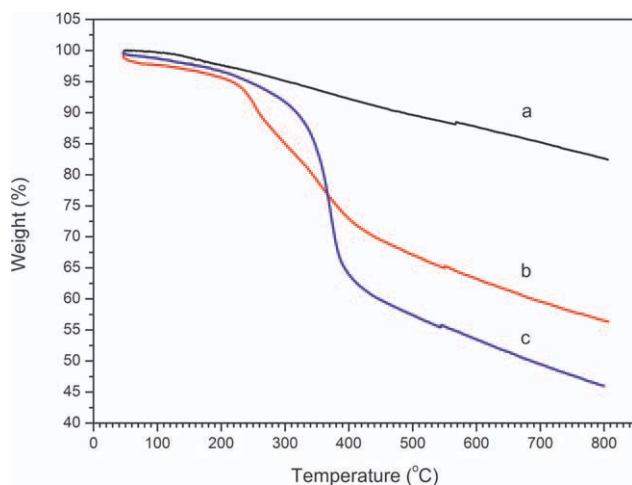


Figure 1. TGA weight loss curves obtained under nitrogen for pristine MWCNTs (a), MWCNT-Jeffamine D400 (b), and MWCNT-Jeffamine D2000 (c). [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

the density of Jeffamine D400 grafted onto MWCNTs ($a = 7.88\%$) is higher than that of Jeffamine D2000 ($a = 2.86\%$), which is probably due to stronger stereo hindrance of Jeffamine D2000 with longer PPO segments than Jeffamine D400.

NMR and FTIR Spectra

The polymer matrix P(AN-GMA) of the nanocomposite used in this study was synthesized by a radical copolymerization, and its composition was determined using $^1\text{H-NMR}$ spectrum as shown in Figure 2. In the spectrum, the signals at 3.27 ppm (d), 2.87 and 2.67 ppm (e) are assigned to the protons of the epoxide group, whereas the peak at 2.16 ppm (f) is attributed to the proton of methine carbon (CH) in AN unit of P(AN-GMA). The peak at 1.98 ppm (b) is due to methylene protons of GMA unit in the copolymer.

The mole content of the GMA unit in the copolymer can be obtained from the EEW titration. The EEW value of P(AN-GMA) is 253, so the mole fraction of the GMA unit $m\%$ in the copolymer can be calculated from the following eq. (2):

$$m = \frac{1}{1 + (\text{EEW} - 142)/53} \times 100, \quad (2)$$

where 142 and 53 are the molecular weights of GMA and AN units, respectively. Thus, the mole fraction of the GMA unit in the P(AN-GMA) copolymer is calculated to be 32%.²²

The gel fraction of the chemically crosslinked nanocomposite based on P(AN-GMA) and Jeffamine-functionalized MWCNTs was determined by extraction of the nanocomposite with THF at 70°C for 24 h (gel fraction (%) = $W/W_0 \times 100\%$ (where W_0 is the weight of polymer before extraction and W is the net

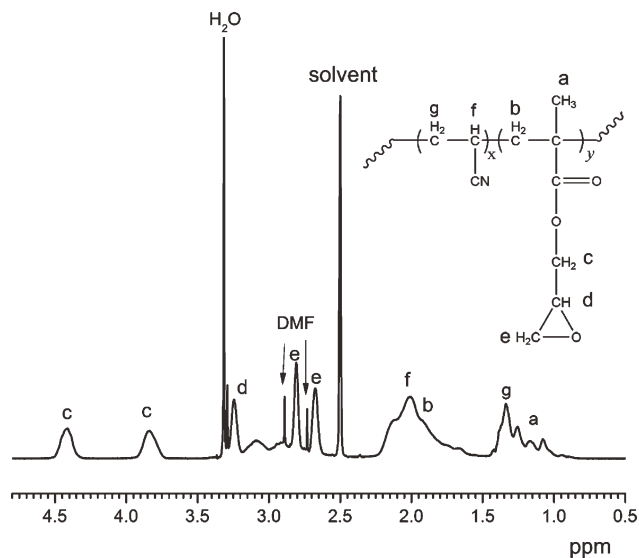


Figure 2. $^1\text{H-NMR}$ spectrum of P(AN-GMA).

weight of polymer after extraction). The results are listed in Table I. It is seen that all the nanocomposites show a value of gel fraction of various degrees, indicating that most of the Jeffamine-functionalized MWCNTs reacted with P(AN-co-GMA) to obtain a crosslinked network structure.

Figure 3 shows the FTIR spectra of MWCNTs, MWCNT-Jeffamine, P(AN-GMA), chemically crosslinked nanocomposites of P(AN-GMA), and Jeffamine-functionalized MWCNTs. An absorption peak attributed to ether group of Jeffamine (1050 cm^{-1}) can be seen in the spectra of Jeffamine-grafted MWCNTs. In the spectra of all the nanocomposites, a characteristic peak of epoxy group can be clearly observed at 908 cm^{-1} , which indicated that the epoxy group in P(AN-GMA) did not react with amine group in Jeffamine-functionalized MWCNTs completely. This may be due to the insufficient active hydrogen in MWCNT-CONH resulting from the small proportion of MWCNT-CONH in the nanocomposite (typically 6 wt %).

SEM Observation

Figure 4 displays the typical SEM images of the cross section of the membranes of the nanocomposites of P(AN-GMA) crosslinked with MWCNT-CONH. The dense membranes were obtained by solution casting. In the pictures, the brighter dots may be attributed to the tips of polymer-functionalized MWCNTs. Apparently, they were dispersed uniformly in the continuous phase of polymer matrix as a result of the formation of strong chemical bonding between the active hydrogen of the functional Jeffamine attached to MWCNTs and the epoxide groups of P(AN-GMA) matrix. As is known, uniform dispersion of MWCNTs fillers and the strong covalent bondings formed between the two components of the nanocomposites may facilitate the load transfer from the polymer matrix to the MWCNTs

Table I. Gel Fraction of Chemically Crosslinked Nanocomposites of P(AN-GMA) and Jeffamine Functionalized MWCNTs

Samples	AG/CNT-D400 (4%)	AG/CNT-D400 (6%)	AG/CNT-D2000 (4%)	AG/CNT-D2000 (6%)
Gel fraction (%)	63.91	76.67	48.69	69.85

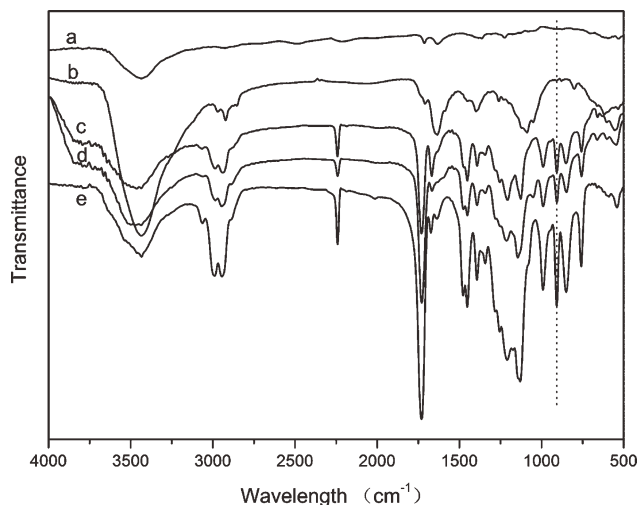


Figure 3. FTIR spectra of MWCNTs (a), MWCNT-Jeffamine (b), AG/MWCNT-D400 (6%) (c), AG/MWCNT-D2000 (6%) (d), and P(AN-GMA) (e).

load,¹⁹ which is supposed to enhance the mechanical properties of the nanocomposites and corresponding GPEs.

Mechanical Properties

The critical strength of the membranes of nanocomposite of P(AN-GMA) and MWCNTs functionalized by the acid or Jeffamine of different molecular weights are presented in Table II, and their stress–strain curves are presented in Figure 5, which are used to evaluate the mechanical properties of corresponding GPEs. It is found that the membrane based on the nanocomposite of P(AN-GMA) and MWCNT-COOH formed by physical blending exhibits a critical stress of 10.99 MPa with an elongation-at-break value of 3.25%. In comparison, the critical stress of the membranes based on the chemically crosslinked nanocomposite of P(AN-GMA) and MWCNTs-CONH is much enhanced, together with the elongation-at-break. The results clearly demonstrate that the formation of strong chemical bondages between the components of the nanocomposites can more effectively improve the mechanical properties as compared to the establishment of physical bindings. Furthermore, it is observed that both the critical strength and elongation-at-break

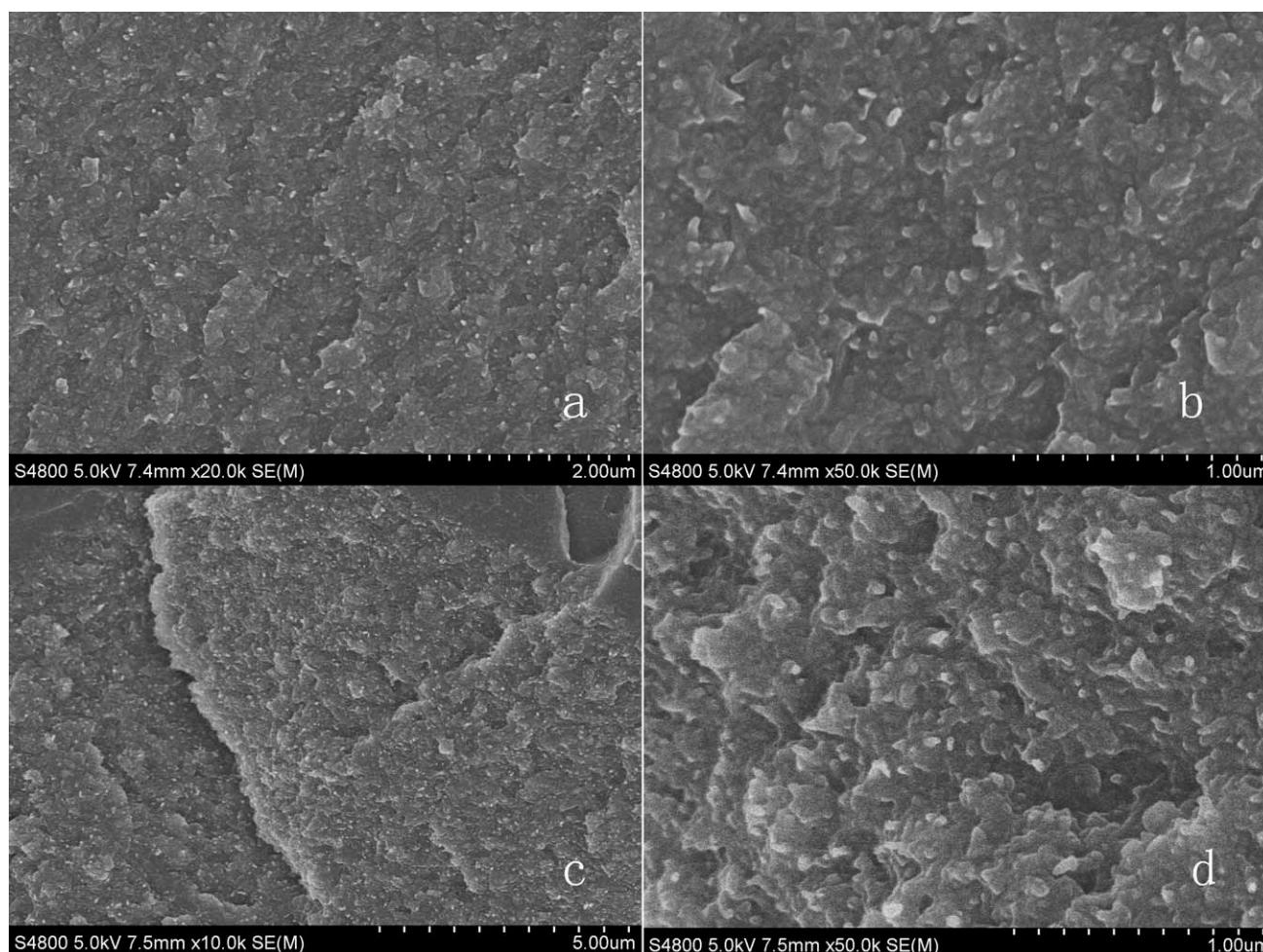


Figure 4. SEM images of the cross-section of membranes of P(AN-GMA) crosslinked with MWCNT-CONH: (a,b) AG/MWCNT-D400 (6%); and (c,d) AG/MWCNT-D2000 (6%).

Table II. Critical Strength, Elongation-at-Break, and Elastic Modulus of the Membranes Based on the Nanocomposites of P(AN-GMA) and MWCNT-COOH or MWCNT-CONH^a

Samples	AG/MWCNT-COOH	AG/MWCNT-D400 (4%)	AG/MWCNT-D400 (6%)	AG/MWCNT-D2000 (4%)	AG/MWCNT-D2000 (6%)
Critical strength (MPa)	10.99 ± 0.25	13.11 ± 0.24	15.22 ± 0.27	28.82 ± 0.33	31.36 ± 1.44
Elongation-at-break (%)	3.25 ± 0.13	5.80 ± 0.24	6.37 ± 0.17	6.26 ± 0.17	8.21 ± 0.13
Elastic modulus	338.42 ± 6.49	226.15 ± 8.53	238.89 ± 10.82	460.12 ± 12.56	382.12 ± 14.14

^aAverage results of five samples.

of the crosslinked nanocomposites of P(AN-GMA) and MWCNT-CONH are higher for samples in which MWCNTs are modified with Jeffamine of higher molecular weights or higher content of MWCNT-CONH fillers are included. It is proposed that the PPO chains of Jeffamine used for modification of MWCNTs may act as a bridge between P(AN-GMA) and MWCNTs after the formation of chemical crosslinking bondages. Thus, longer flexible PPO chains in Jeffamine of higher molecular weights may be more effective in transferring the load from P(AN-GMA) to MWCNTs, which in turn leads to enhanced mechanical properties for the nanocomposite. Similarly, higher content of MWCNT-CONH facilitates the load transfer and better mechanical properties can be obtained. The elastic modulus of crosslinked AG/CNT-D400 is lower than the physical blending samples, whereas the AG/CNT-D2000 is much higher. This may relate to better interfacial compatibility of Jeffamine D2000 than Jeffamine D400 in the composites of cross-linked polymer and MWCNTs.

Thermal Analysis

DSC traces (second heating runs) of the nanocomposites of P(AN-GMA) and functionalized MWCNTs are displayed in Figure 6, and the results of thermal analysis are summarized in Table III.

It can be seen that the glass transition temperature (T_g) of P(AN-GMA)/MWCNT-D400 (4%) (80.9°C) is a bit higher than

that of P(AN-GMA)/MWCNT-COOH nanocomposite membrane ($T_g = 79.3^\circ\text{C}$). With the increase in MWCNT-D400 (or MWCNT-D2000), T_g becomes lower. For the chemically cross-linked nanocomposites, T_g is affected by the combined effect of the degree of crosslinking and the segmental mobility of Jeffamine grafted onto MWCNTs. It is true that Jeffamine-modified MWCNT-CONH crosslinked with P(AN-GMA), leading to strong chemical bondages. In comparison, only weaker physical binding was established between the nanocomposite of acid-treated MWCNT-COOH and P(AN-GMA). However, it is to be noted that the long PPO chains of Jeffamine attached to MWCNTs are flexible and their easy segmental mobility usually result in the reduction in T_g . More work is necessary to have a deep understanding of the effect of the composition and structure of the nanocomposites on their T_g .

Electrochemical Properties

The effect of temperature on conductivity of the GPEs based on P(AN-GMA) blended with carboxylated MWCNTs and P(AN-GMA) crosslinked with MWCNTs functionalized with Jeffamine of different molecular weights has been investigated and $-\lg \sigma$ versus $1000/T$ is depicted in Figure 7. It is found that $\lg \sigma$ changes linearly with $1/T$ for all the specimens, indicating an Arrhenius-like behavior of conductivity variation with temperature according to eq. (3).

$$\sigma = \sigma_0 \times \exp(-E_a/KT), \quad (3)$$

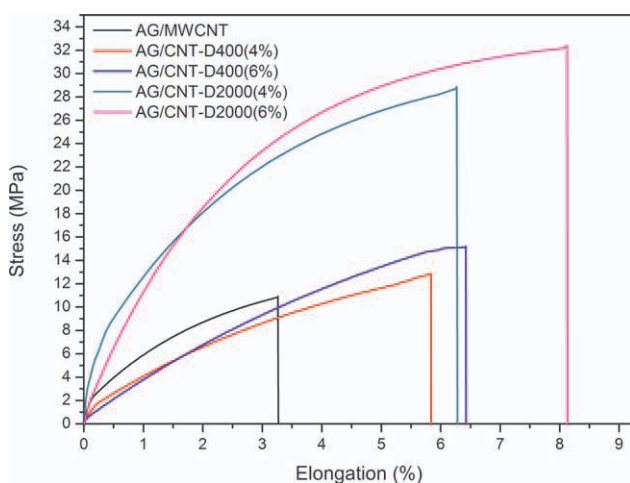


Figure 5. Stress–strain curves of polymer/MWCNT nanocomposites. [Color figure can be viewed in the online issue, which is available at [wileyonlinelibrary.com](http://www.wileyonlinelibrary.com).]

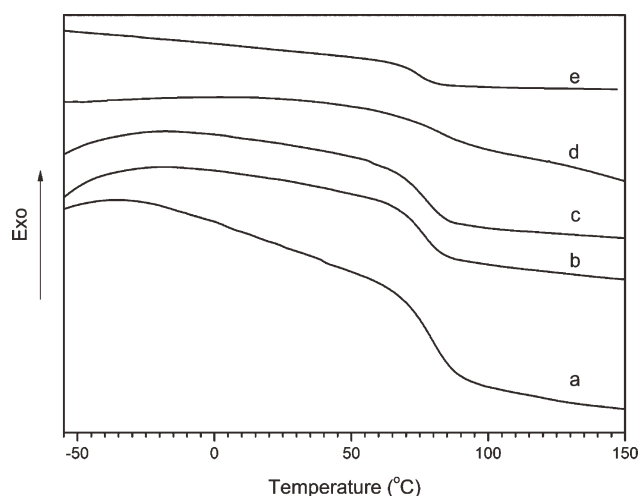


Figure 6. DSC thermograms of AG/MWCNT-COOH (a), AG/MWCNT-D400 (4%) (b), AG/MWCNT-D400 (6%) (c), AG/MWCNT-D2000 (4%) (d), and AG/MWCNT-D2000 (6%) (e).

Table III. T_g of P(AN-GMA)/MWCNT-COOH and P(AN-GMA)/MWCNT-CONH

Samples	AG/CNT-COOH	AG/CNT-D400 (4%)	AG/CNT-D400 (6%)	AG/CNT-D2000 (4%)	AG/CNT-D2000 (6%)
$T_g/^\circ\text{C}$	79.3	80.9	74.5	77.1	77.3

where σ_0 is a constant; E_a is the activation energy; and K is the Boltzmann constant.

From Figure 6, it can be seen that the GPE based on the nanocomposite of P(AN-GMA)/MWCNT-COOH with only physical binding interactions exhibited the lowest ionic conductivity over the temperature range investigated. In comparison, the ionic conductivity of the GPEs based on P(AN-GMA)/MWCNT-CONH with strong chemical bondages were enhanced. This may be related to the PPO chains between P(AN-GMA) and MWCNTs, whose segmental movement could facilitate the mobility of lithium ions. Moreover, it is observed that the GPEs based on the chemically crosslinked nanocomposite of P(AN-GMA) and MWCNTs functionalized with Jeffamine of higher molecular weights exhibited lower ionic conductivity over the range of tested temperature. Among the specimens investigated, the GPE based on AG/CNT-D400 (6%) exhibited the highest ionic conductivity over the range of 15–75°C (3.39×10^{-4} S cm^{-1} at 25°C). The results show that P(AN-GMA)/MWCNT nanocomposite in which chemical crosslinking reactions occurred exhibited not only enhanced mechanical properties but also higher ionic conductivity as compared with the nanocomposite with only physical binding interactions between the components, and prove that chemical crosslinking modification can be an effective method for improving the properties of polymer composites for GPEs.

CONCLUSIONS

Novel nanocomposite GPEs can be obtained based on P(AN-GMA) crosslinked with α,ω -diamino PPO (Jeffamine)-function-

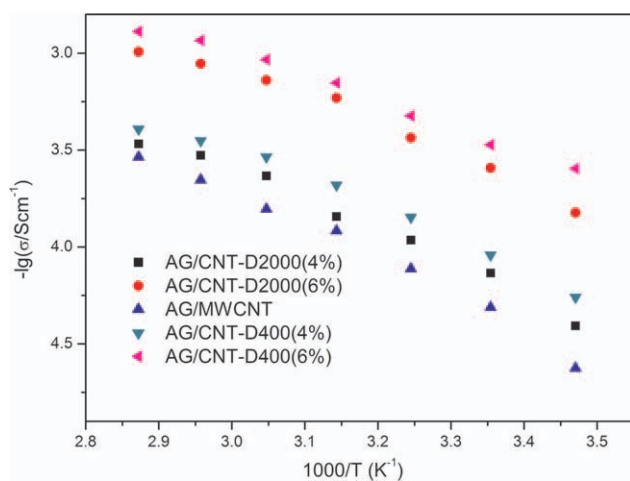


Figure 7. Dependence of conductivity on the reciprocal of temperature for the nanocomposites of P(AN-GMA) with MWCNT-COOH and MWCNT-CONH. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

alized MWCNTs. The Jeffamine-modified MWCNTs were uniformly dispersed in the polymer matrix in the nanocomposite. The chemically crosslinked nanocomposites of P(AN-GMA) and Jeffamine-modified MWCNTs exhibited much improved mechanical properties compared with that physically blended P(AN-GMA) and acid-treated MWCNTs. The ionic conductivity of the GPEs based on the nanocomposite of P(AN-GMA) and functionalized MWCNTs obeyed an Arrhenius behavior over the range of 15–75°C. The GPEs based on chemically crosslinked nanocomposite of P(AN-GMA) and Jeffamine-modified MWCNTs exhibited higher ionic conductivity than that based on physical blend of P(AN-GMA) and acid-treated MWCNTs. The GPE of P(AN-GMA) crosslinked with 6 wt % of Jeffamine D400-modified MWCNTs exhibited the highest conductivity of about 3.39×10^{-4} S cm^{-1} at 25°C.

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