Mechanical Properties and Ionic Conductivity of Gel Polymer Electrolyte Based on Poly(vinylidene-fluoride-*co*-hexafluoropropylene)

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ABSTRACT: Gel polymer electrolytes were prepared from poly(vinylidene-fluoride-*co*-hexafluoropropylene) (PVH), which had higher mechanical properties and a higher dielectric constant ($\epsilon = 8$–13) than other polymeric matrixes. The mechanical properties and ionic conductivities were investigated as a function of the blend ratio of the electrolyte solution and polymer matrix. Ethylene carbonate/γ-butyrolactone and lithium triflate were used as the solvent and salt, respectively. The mechanical properties such as the tensile strength, tensile modulus, compression modulus, and dynamic shear modulus were evaluated. The highest ionic conductivity was 1.09×10^{-3} S/cm at 25°C for 40 wt % PVH against the electrolyte solution. The tensile strength, tensile modulus, and compression modulus were increased with increasing PVH content and they all abruptly changed between 70 and 80 wt % PVH. The dynamic shear moduli showed behavior typical of a gel and varied with the shear strain amplitude or applied frequencies. © 2001 John Wiley & Sons, Inc. J Appl Polym Sci 81: 948–956, 2001

Key words: gel polymer electrolyte; mechanical property; poly(vinylidene-fluoride-*co*-hexafluoropropylene); ionic conductivity; 3-dimensional network

INTRODUCTION

In recent years it has become possible to share much information without regard for time and space because of the appearance of portable communication devices such as cellular phones, personal computers, notebook computers, and camcorders. Therefore, lithium polymer secondary batteries are of the interest as a power source because of their rechargeability, high energy density, ability to be leak proof, lack of environmental pollution, and broad electrochemical stability.[1–3] Generally, suitable polymer electrolytes for a lithium secondary battery can be classified as solid polymer electrolytes (SPEs), gel polymer electrolytes (GPEs), and hybrid polymer electrolytes (HPEs). Among them, SPEs composed of polymers with a polar group (O, N) in the main chain and lithium salt were extensively investigated since the ionic conductivity of a poly(ethylene oxide) (PEO)–salt complex was reported by Wright[4] and Armand et al.[5] However, this system had a limitation on practical applications because of low ionic conductivity resulting from the high crystallinity of PEO at room temperature. Accordingly, many approaches were adopted to reduce the crystallinity of PEO-based electrolytes and in-

crease the segmental mobility of the host polymer through copolymerization,[6–8] grafting,[9,10] and network formation.[11–14]

On the other hand, GPEs with high ionic conductivity at room temperature and an easy manufacturing method were developed more recently. In GPEs the polymeric materials play a role as a matrix and in lithium ion transport through an organic solvent or mixture of solvents that have low viscosity, a high dielectric constant, low volatility, and excellent electrochemical stability, considering the polymer–solvent compatibility. In many cases the polymeric materials that were used were polyacrylonitrile,[15,16] poly(vinyl chloride),[17] poly(methyl methacrylate),[18,19] and poly-(vinylidene fluoride) (PVDF).[20–26] Tsuchida et al. reported PVDF-based GPEs with ionic conductivities in the region of 10^{-5} S/cm at room temperature.[20,21] A suitable composition for their electrolytes was PVDF/propylene carbonate/lithium perchlorate with a molar ratio of 50/30/20, and lithium perchlorate was homogeneously dispersed in the hybrid film with this composition as revealed by an X-ray diffraction measurement. Abraham et al. also reported that GPE films comprising PVDF/propylene carbonate/lithium triflate had wide dimensional stability and showed 10^{-3} S/cm ionic conductivity at 30°C.[22] Voice et al. made thermoreversible GPEs with an ambient ionic conductivity of about 10^{-3} S/cm by using organic solvents such as DMF, DMSO, and tetraethylene glycol dimethyl ether.[23] Croce et al. reported that the choice of salt was more important than the choice of organic solvent as found with lithium NMR research.[24] In addition, Whang and Lu reported that the ionic conductivities of PVDF-based GPEs displayed a stronger dependence on the content of chlorine-terminated poly(ethylene ether), which was used as plasticizer, than did the PEO-based GPE.[25]

As in the studies above, most of the research on GPEs focused on their ionic conductivities at room temperature, and few studies on their mechanical properties were performed up to the present time. However, the GPEs that were manufactured in very thin films with thicknesses of 15–20 μm simultaneously acted as a transport for lithium ions, a separator, and a binder between the anode and cathode. Thus, GPEs should have enough mechanical properties to support the pressure induced by stacking of electrodes or stress coming from morphological deformation of the electrode during repeated charge–discharge. In addition, the effect of the mechanical properties of GPE films on battery efficiency has in-

creased toward research, processing development, and manufacturing steps.

In this research, GPEs were prepared from poly(vinylidene fluoride-co-hexafluoropropylene) [P(VDF-co-HFP) or PVH], which had higher mechanical properties and a higher dielectric constant ($\epsilon = 8$–13) than other polymeric materials, to study the relationship between the mechanical properties and ionic conductivity. The tensile and compression properties, dynamic shear modulus, ionic conductivities, and thermal properties of the GPEs were investigated as a function of the content of PVH and the optimum composition ratio of the PVH/electrolyte solution (ES).

EXPERIMENTAL

Materials

The PVH (MW 400,000, pellet, Aldrich) as a polymer matrix was dried *in vacuo* at 50°C for 24 h before use. Ethylene carbonate (EC) and γ-butyrolactone (γ-BL) were used as a carrier for the lithium ions and as a plasticizer in the GPEs (Aldrich); they were dehydrated over molecular sieves before use. Lithium triflate (LiCF$_3$SO$_3$) as a lithium salt was dried *in vacuo* at 80°C for 48 h prior to use, and acetone (99.0%, Showa Chemical Co.) was used as a cosolvent.

Electrolyte solutions with molar concentrations of 0.25, 0.5, 0.75, 1.0, 1.5, and 12.0M were prepared by mixing the solvents (EC/γ-BL, 1:1 mole ratio) and the LiCF$_3$SO$_3$ lithium salt.

The PVH pellet (not the powder form) was completely dissolved in acetone with stirring for 2 h at room temperature and then further stirring at 50°C for 30 min in a 100-mL flask. The lithium triflate was dissolved in an appropriate amount of the solvent mixture (EC/γ-BL, 1:1 mole ratio), resulting in a 1M lithium ES. A transparent and homogeneous polymer ES was obtained by mixing the 1M lithium ES and PVH polymer solution together.

Preparation of GPEs for Characterization

The polymer ES for the ionic conductivity measurements was dropped on a slide glass and cast using a scalpel. Freestanding GPE films with thicknesses ranging from 0.1 to 0.2 mm were formed during the evaporation of acetone.

For the mechanical property measurements the polymer ESs were poured into a Teflon casting mold, which had dimensions of 85 $\times$ 25 $\times$ 20

Table I   Compositions of Gel Polymer Electrolytes Containing PVH

Sample Name	PVH Content vs. 1*M* ES (wt %)	Content of Each Component (Total wt %)	
		PVH	1*M* ES
PVH40	40	28.6	71.4
PVH45	45	31.0	69.0
PVH50	50	33.3	66.7
PVH60	60	37.5	62.5
PVH70	70	41.2	58.8
PVH80	80	44.4	55.6

PVH, Poly(vinylidene-fluoride-*co*-hexafluoropropylene); ES, electrolyte solution.

mm. When the acetone was evaporated thoroughly, translucent, freestanding GPE films were formed, which had a thickness range of 0.3–2.3 mm according to the measurement method. The thicknesses of the GPE films were regulated by the amount of solution poured into the Teflon mold. The codes for all samples prepared in this research are listed in Table I. The numbers (40, 45, 50, 60, 70, 80) in the code stand for the weight percent of PVH versus the 1*M* ES.

The DSC measurements of the GPE films were studied using a Perkin–Elmer DSC7 equipped with a cooling apparatus in a temperature range of −70 to 60°C at a scanning rate of 10°C/min under a dry helium flow. Each sample was dried *in vacuo* for 24 h before measurement and then sealed in an aluminum pan. The TGA of the GPE films were performed with a Perkin–Elmer TGA7 in a temperature range of 30–600°C at a scanning rate of 10°C/min.

The ionic conductivities of the GPE films were evaluated by the complex impedance method using a frequency response analyzer (SI 1260, Schlumberger Co.) equipped with a Mettler hot stage; frequencies ranging from 1.0 to 1.0×10^6 Hz were used. The bulk resistance of the samples was measured with a temperature range of 25–60°C and a constant applied signal amplitude of 0.1 V using stainless steel (SUS304) as a blocking electrode. The ionic conductivity (σ) was calculated from the bulk electrolyte resistance value (R_b) found in the complex impedance diagram according to the following equation:

$$\sigma = \frac{l}{R_b \cdot A} \qquad (1)$$

where l is the length and A is the absorbance.

The static mechanical properties of the GPE films were evaluated from stress–strain tests using an Instron model 4465 testing device equipped with an extensometer and interfaced to an IBM-compatible computer for data collection. Tensile tests were carried out under a 10-N load cell at a strain rate of 50 mm/min. Compression tests were conducted under a 5-kN load cell at a crosshead speed of 10 mm/min, and the GPE samples were 25 mm in diameter and approximately 2 ± 0.3 mm in thickness. All measurements were performed 5 times for each sample for an average.

Using samples with the same geometry as the compression test, the dynamic mechanical properties of the GPE films were measured with an ARES instrument (Rheometrics Co.) in a nitrogen atmosphere over a frequency range of 0.05–100 Hz and a temperature range of 30–60°C. In the strain sweep the strain range was 1–20% at room temperature.

RESULTS AND DISCUSSION

Ionic Conductivity

From a general point of view, the ionic conduction in GPEs depends on various factors. These are the mobility and number of carrier ions in the ES, as well as the availability of a connecting polar domain as the conduction pathway.[3] By optimizing these factors, high ionic conductivity may be realized. The ionic conductivity in liquid ES is simply defined in eq. (2).

$$\sigma = \sum n_i e \mu_i \qquad (2)$$

where n_i is the number of ions of species i, e is the charge, and μ_i is the mobility of the i ions.

Equation (2) shows that the ionic conductivity may vary if the mobility of the charge carriers varies and the predominant charged species changes. Figure 1 shows the behavior of the ionic conductivity with the concentration of the ES (EC/γ-BL/LiCF$_3$SO$_3$). The maximum ionic conductivity was found at 1*M* ES. This dependence is commonly explained in terms of ion–ion interactions. As the concentration of an ES is increased, the mutual distance between ions decreases with the increasing number of charged carriers until ion–ion interactions become significant. Therefore, the higher the concentration, the stronger the ion–ion interaction, resulting in a change of free ions to ion pairs or to the formation of higher
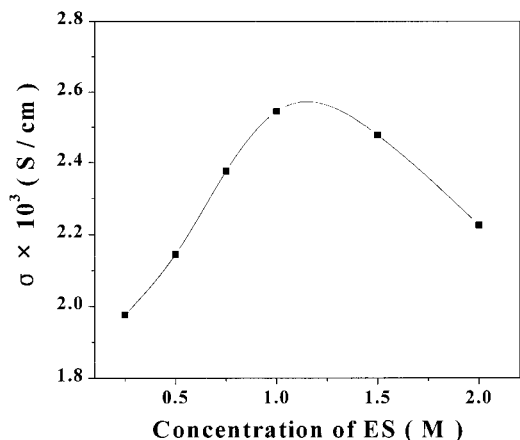
Figure 1 The ionic conductivity versus the concentration of the lithium salt solution electrolyte.

aggregates. Consequently, the ionic conductivity decreases as a result of the decrease of mobility, as well as the number of charge carriers. Thus, in the EC/γ-BL/LiCF$_3$SO$_3$ we confirmed that 1M ES was the optimum concentration for the GPE that is discussed in this article. The conduction pathway, which charged ion transport effectively, was taken into consideration. In the GPE structure the conductive phase (ES) must be continuous in the direction of the thickness because lithium ions need to be passed from one side of the GPE film to the other.[27] The supporting phase (polymer matrix) must also be continuous in all the directions. The ionic conductivity changes of the GPEs as a function of polymer matrix contents with various temperatures are shown in Figure 2.
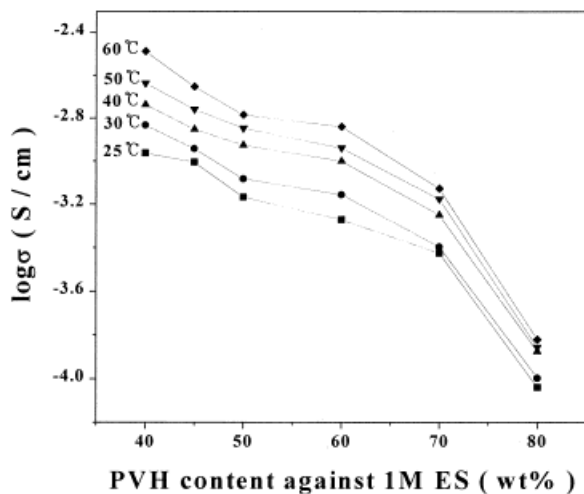

Figure 2 The ionic conductivity as a function of PVH contents versus the 1M ES solution at various temperatures.
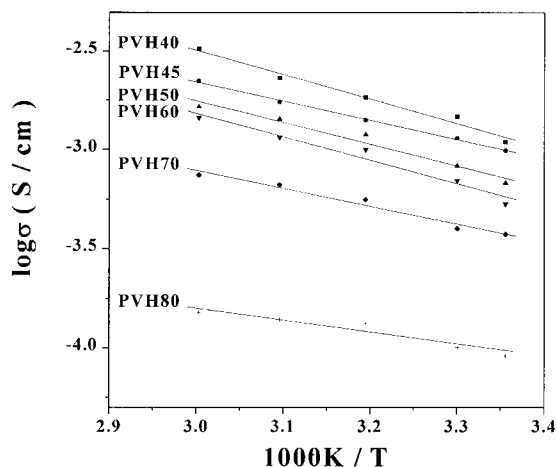

Figure 3 The temperature dependence on the ionic conductivity for various PVH contents versus the 1M ES.

The conductivity decreased with the increasing content of PVH, and there was an especially dramatic decrease between PVH70 and PVH80. In our system the ionic conductivity mainly depended on whether or not many continuous conduction pathways were formed because the concentration of the ES was held constant as 1M. Accordingly, as the amount of polymer matrix was increased and the amount of ES was decreased, the ionic conductivity decreased because of a decrease of the number of conduction pathways. In the dramatic drop between PVH70 and PVH80, when the polymer matrix was contained beyond a certain limitation, the extent of conduction pathways was markedly decreased, leading to a sudden decrement of ionic conductivity.

Figure 3 describes the changes of the ionic conductivity of GPE films in the temperature range of 25–60°C. Because linear relations were observed in all cases, the ionic conductivity followed the Arrhenius behavior in eq. (3). This meant that there was no phase transition in the polymer matrix and the ion conduction mechanism of the GPE films studied in our research was not by dynamic configuration of the polymer matrix but was by the conduction path that was formed by the mixture of solvents.

$$\sigma = \sigma_0 \exp(-E_a/RT) \qquad (3)$$

where σ is the ionic conductivity, σ_0 is a constant, E_a is the activation energy, R is the gas constant, and T is the temperature.

Thermal Properties

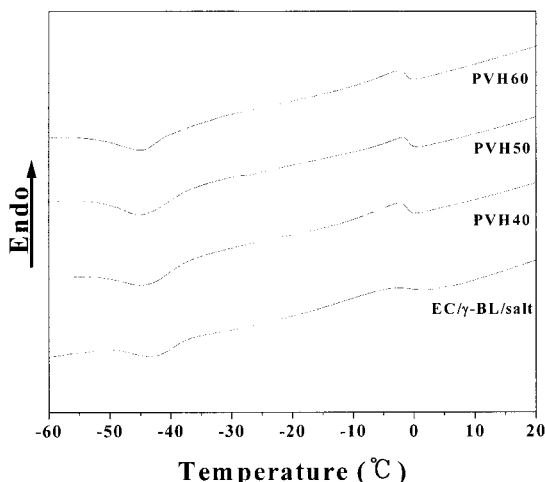The DSC thermograms of the GPE films and lithium ES are illustrated in Figure 4. The recrystal-

Figure 4 DSC thermograms of gel electrolyte films and 1*M* ES.

lization peaks and melting peaks of the GPE films were found at around -43 and $-3°C$, respectively. These peaks were attributed to the crystal complexes of the mixture of solvents and salt below $-3°C$, which was confirmed by the same result for the mixture of solvents and salt in the DSC thermograms.[28] Because these crystalline phases in the conducting medium reduced the ionic conductivity, this suggested that the ionic conductivity would be abruptly changed at $-3°C$, corresponding to the melting point of the crystal complexes of the mixture of solvents and salt.[20]

Figure 5 shows TGA thermograms of the GPE films. The GPEs decomposed in the same way. All the films exhibited a relatively small weight loss of <4% in the temperature range of 30–100°C, which was presumably due to the evaporation of residual moisture absorbed when the samples were loaded and the infinitesimal evaporation of the solvent mixtures. From 100 to 270°C weight loss abruptly occurred that was attributable to the evaporation of the ES. Thus, these GPE films were stable up to 100°C, which was higher than the operating temperature of the lithium polymer cell (50–70°C). Above 270°C (i.e., after complete evaporation of the ES) no weight loss was observed until irreversible decomposition of PVH began at around 360°C.

Mechanical Properties

A gel is expressed as a soft, solid, or solidlike material that consists of more than two components; one of these is a liquid, which is present in a substantial quantity. This gel can be divided into two main classes: chemically crosslinked ma-

terials and polymer networks formed through physical aggregation, which is predominantly disordered, but with regions of order. In a physical gel the crosslinks do not act at a point on the chain as do covalent crosslinks; instead they involve more extended "junction zones" or even particulate structures. In this section we comment on the various mechanical properties of a physical gel including the substantial quantity of lithium ES.

Static Mechanical Properties

Generally, tensile tests are used to investigate the mechanical properties of GPE films. However, because GPE films play the roles of an electrolyte and a separator between electrodes under pressure in the battery manufacturing process, the compression property is also important factor in terms of the life cycle and battery manufacturing process. Three significant evaluations of GPE films were obtained from a stress–strain curve.

The stress–strain curves of all the prepared GPE films are presented in Figure 6. All measured samples showed rubberlike behavior because PVH has the properties of a thermoplastic elastomer when used as a polymer matrix. It was noticed that PVH40 possessed a 1.1 MPa elongation at break value (strain) of 213%, although it contained the minimum content of polymer matrix (ca. 28.6 wt %) among the GPE films measured. These values were higher or similar in comparison with other research.[22,27,29] The strain amplitude was increased with increasing content of PVH, except between PVH70 and PVH80. It was for this reason that as the content of the PVH
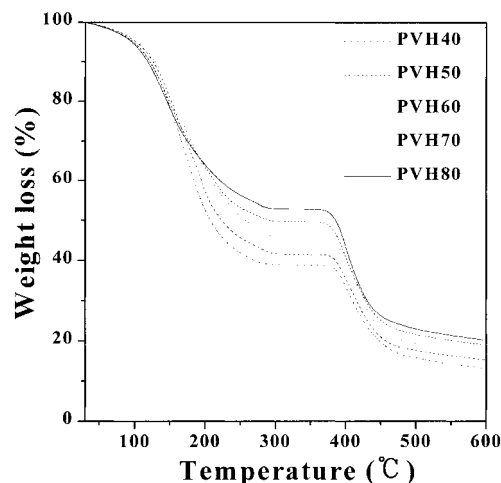

Figure 5 TGA thermograms of gel electrolyte films as a function of the PVH content versus the ES content.
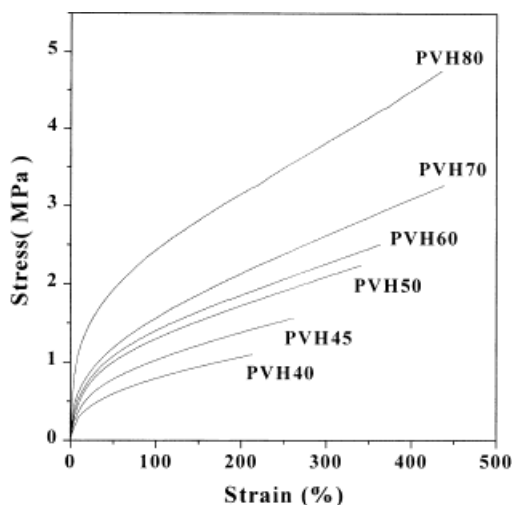
Figure 6 Tensile stress–strain curves as a function of the PVH contents versus 1M ES at room temperature.

Figure 8 The compression modulus and ionic conductivity as a function of the PVH contents versus 1M ES at room temperature.

in the GPE was augmented, the formation of a 3-dimensional (3-D) network of the gel was improved, which led to a continuous phase formation of the polymer matrix.[30,31] Figure 7 shows tensile modulus as a function of content of PVH. The tensile modulus was also steadily increased as the content of PVH was increased, but it showed a rapid change between PVH70 and PVH80. This was the inverse of the trend of the ionic conductivity, as shown in Figure 2. It could also be explained as the formation of the 3-D network of the gel. The concentration of polymer chains per unit volume increased as the PVH content increased; this led to the development of
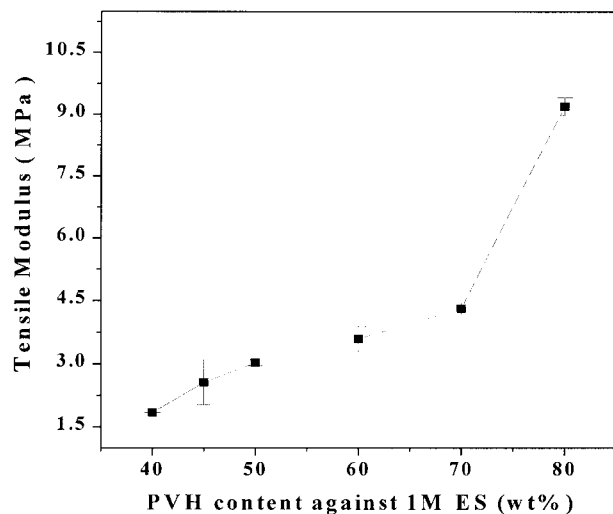

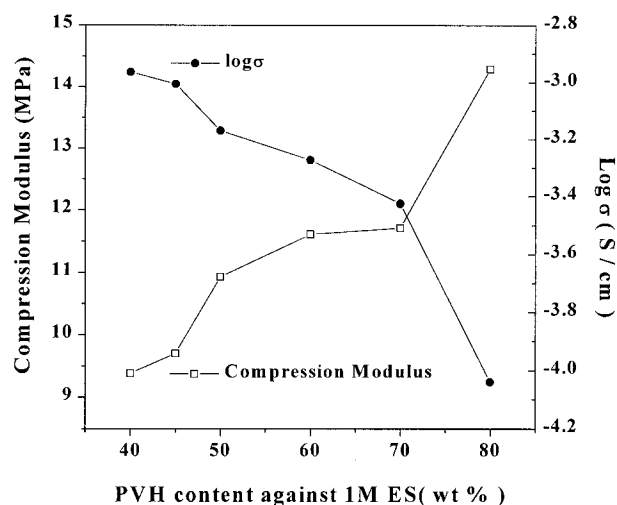**Figure 7** Tensile modulus variations with the PVH contents versus 1M ES at room temperature.

junction zones of the physical gel, which affected the increase of the total stress of the GPE but had less affects on that of the strain. The increases of the tensile strength and modulus corresponding to this result are shown in Figures 6 and 7, respectively. In addition, we thought that there were considerable developments of the 3-D network of the gel between PVH70 and PVH80.

Figure 8 is a plot of the variation in compression modulus with the content of PVH. In general, the modulus determined in the compression test was higher than that determined in the tensile test. This could be a reflection of both of the different stress fields that were applied, but it was more likely to be due to the occurrence of different molecular or small-scale processes that can occur in the different tests.[32] Besides, in case of the GPE the determination of the compression modulus required consideration of the movement of ES entrapped under the applied load. The applied load was supported partly by the ES and partly by the solid network when the gel was compressed. As the complete gel was compressed, the ES could not instantly flow out of the network and perform as if it were incompressible, so the load had to be sustained until the flow of ES allowed the pressure in the ES to drop to zero and the load was transferred entirely to the solid network.[33] Accordingly, as the content of PVH decreased, the degree of formation of the 3-D network decreased and the ES promptly flowed out of the GPE, resulting in a decrement of the compression modulus. It was also thought that the quite large variation between PVH70 and PVH80 resulted from

the considerable change of the 3-D network of the gel at this boundary. According to these results, the PVH content in the GPE was the dominant factor responsible for its mechanical properties. Consequently, it was suggested that PVH70 was the optimum composition ratio (see Table I) because of its relatively good ionic conductivity (0.37×10^{-3} S/cm) and excellent mechanical properties.

Dynamic Mechanical Properties

For purely entangled systems, such as a polymer melt, as the frequency is decreased there is a crossover in the storage modulus (G') and loss modulus (G''). At very low frequencies they flow as high viscosity liquids in the terminal zone. By contrast, true gels should not show such an effect and the G' and G'' for a rubbery gel (i.e., above its glass transition) are parallel and largely frequency insensitive. In addition, the gel can be divided into the two main classes of chemically crosslinked materials (including bulk elastomer) and physically crosslinked polymer networks formed through predominantly disordered physical aggregation, but with a region of order.

The variations of the G' and G'' of PVH60, PVH70, and PVH80 with the strain amplitude at constant frequency (0.05Hz) are shown in Figure 9. At low strain amplitude (~1%) the G' dominated over the G'', which meant that the sample could be described as elastic. However, the G' markedly decreased with increasing strain amplitude and crossover occurred; consequently, the G'' dominated the G' at high strain amplitude. This can be explained by the fact that the viscosity factor was stronger than that of the elastic at high strain amplitude because of the breaking of the 3-D network of GPE in some ways, although this breakdown was spontaneously recoverable when the applied strain was removed.[23] In the PVH 60 and PVH70 these crossovers occurred at strain amplitudes of 10 and 12%, respectively, while the crossover of PVH80 appeared at 16%, which showed about a twofold greater difference than that between PVH60 and PVH70. This result reflected that PVH80 was a stronger gel than PVH70; again, there was considerable development of the 3-D network of the physical gel between PVH70 and PVH80.
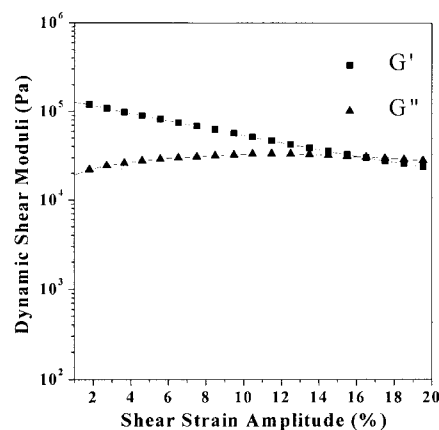
The variations of the G' and G'' of PVH40 are plotted as a function of frequency at various temperatures at low strain amplitude (1%) in Figure 10. The figure shows that the G' and G'' remained
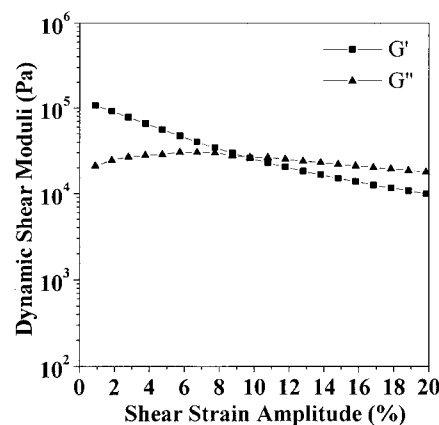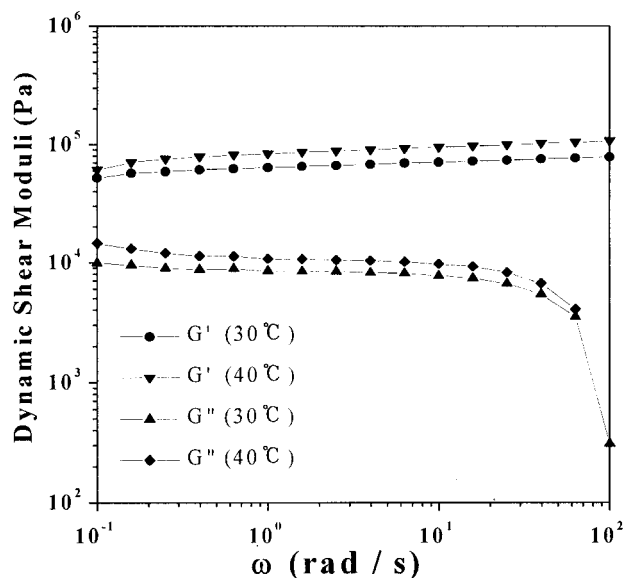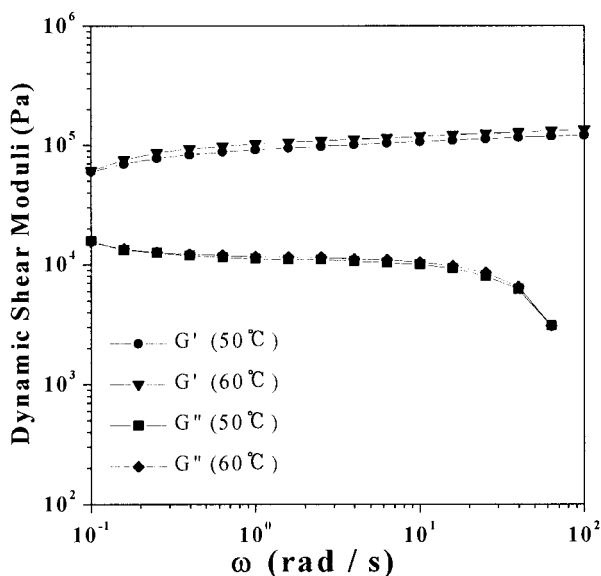
(a) PVH60
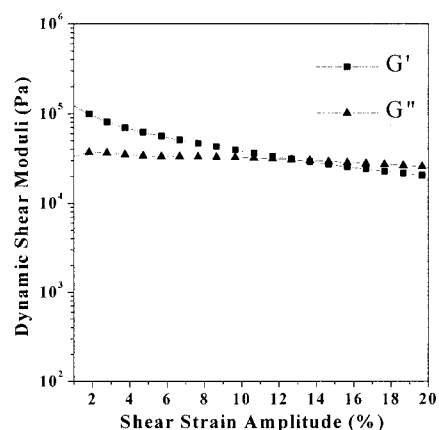
(b) PVH70

(c) PVH80

Figure 9 The variation of the dynamic shear modulus for GPE films at room temperature with shear strain amplitudes at 0.05 Hz: (a) PVH60, (b) PVH70, and (c) PVH80.

approximately constant and G' dominated G'' over the frequency range measured. This was in good accord with the definition of the gel given by

(a) 30℃, 40℃

(b) 50℃, 60℃

Figure 10 The variation of the dynamic shear modulus for PVH40 with frequencies at 1% shear strain amplitude at (a) 30, 40, and (b) 50°C.

Almdal et al., which was that the gel was a material with a G' that exhibited a plateau extending to times on the order of seconds and a G'' that was smaller than G' in the plateau region.[34] Also, a noticeable variation of the gel in the temperature range measured was not found, although the weakest GPE film was among the samples prepared in this research.

CONCLUSIONS

We prepared PVH-based GPEs with excellent mechanical properties and investigated their mechanical properties and ionic conductivities with the content of PVH versus the content of ES. The maximum ionic conductivity was found at a 1M concentration for the lithium ES as the result of a decrease of the number of charge carriers and their mobility. In the GPE films the highest ionic conductivity was 1.09×10^{-3} S/cm for PVH40. This mainly depended on whether or not continuous conduction pathways were formed because the concentration of ES was held constant at 1M. The static mechanical properties such as the tensile strength, tensile modulus, compression modulus, and the ionic conductivity were abruptly changed between PVH70 and PVH80. The dynamic mechanical properties showed behavior typical of a gel; especially the G' and G'' markedly varied with the strain amplitude resulting from the reversible 3-D network of the GPE. Therefore, it was suggested that the developments of 3-D network structures of the GPEs were responsible for our results, and PVH70 was the optimum composition ratio because of its relatively good ionic conductivity and excellent mechanical properties.

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