Available online at www.sciencedirect.com

SCIENCE DIRECT®

Polymer 46 (2005) 11499–11504

polymer

www.elsevier.com/locate/polymer

Preparation of thermally stable polymer electrolytes from imidazolium-type ionic liquid derivatives

Hiromitsu Nakajima [a,b], Hiroyuki Ohno [b,*]

[a] Oleo and Speciality Chemicals Research Laboratory, NOF Corporation, Kawasaki, Kanagawa 210-0865, Japan
[b] Department of Biotechnology, Tokyo University of Agriculture and Technology, Koganei, Tokyo 184-8588, Japan

Received 14 April 2005; received in revised form 30 August 2005; accepted 1 October 2005
Available online 20 October 2005

Abstract

Thermally stable polymer electrolytes based on ionic liquids were prepared and analyzed. Mono-functional ionic liquid monomers, ionic liquid cross-linkers, and ethylimidazolium-type ionic liquid salts were mixed and polymerized. The ionic liquid-type cross-linkers were effective to prepare thermally stable polymer films. In particular, the copolymerization of cross-linker and ethylimidazolium-type ionic liquid monomers were used to make polymer electrolytes with high ionic conductivities. The copolymerization in ethylmethylimidazolium bis(trifluoromethanesulfonyl) imide gave a transparent film showing no thermal degradation up to 400 °C.

Keywords: Ionic liquid; Polymer electrolyte; Thermal stability

1. Introduction

Various ionic liquids (ILs) have been developed and studied since the first air-stable ionic liquid was reported in 1992 [1]. The applications of ILs that have attracted the most attention are green solvents [2] and battery electrolytes [3]; however, they have many other potential uses including those in lithium ion batteries [4], fuel cells [5], solar batteries [6], capacitors [7], and actuators [8]. In addition, fixations of ILs are expected to be a great help for making thin layers and miniature ionic devices. For such devices, both high ionic conductivity and thermal stability are required. It is generally difficult to prepare thermally stable organic system, especially for the complex systems with several functional groups, because they start decomposing at the weakest bond. Ether bonds are frequently used for the potential polymer electrolytes due to following characteristics such as suitable dipole moment for pseudo-solvation to ions and low glass transition temperature (T_g). Former enhances the dissociation of added salts and the latter helps to transport these dissociated ions. However, the ether bond is not thermally stable and gradual degradation occurs at around 160 °C. Therefore, new thermally stable polymer systems are expected for fast ion conduction without ether groups. Ionic liquids (ILs) are generally thermally stable and highly ion conductive. The decomposition temperature of some ILs is reported to be above 400 °C [9]. It is easy to understand that the polymerization of ILs is believed to give thermally stable polymer electrolytes. The initial step of this purpose is the mixing of ordinary polymer and ionic liquids. Thus prepared mixture is called ionogels. An ionogel is prepared by either polymerization of various monomers in the presence of ILs or a simple mixing of polymers with ILs. The important factor for various applications is the compatibility between ILs and polymers. In spite of facile preparation, thermal stability of the ionogel is generally not so favorable because of poor thermal stability of the component polymers. In previous studies, we examined the polymerization of various ionic liquid vinyl monomers to prepare polymers such as polycation [10], polyanion [11], polymer complex, copolymer [12], and poly(zwitterions) [13]. However, due to a decrease in the number of mobile ions and considerable elevation of T_g, polymerization of ionic liquid monomers significantly decreases the ionic conductivity. To overcome this problem, we investigated ionic-liquid-type polymer brushes having a flexible spacer between the polymerizable group and the imidazolium cation. Thermal stability up to 390 °C was attained in this system [14,15].

In the present study, we synthesized new cross-linking monomers containing an ionic liquid moiety, and some ionic liquid monomers were polymerized in the presence of this ionic

---

* Corresponding author. Tel./fax: +81 42 388 7024.
E-mail address: ohnoh@cc.tuat.ac.jp (H. Ohno).

0032-3861/$ - see front matter © 2005 Elsevier Ltd. All rights reserved.
doi:10.1016/j.polymer.2005.10.005

liquid type cross-linker to improve both thermal stability and mechanical strength.

2. Experimental

2.1. Materials

We used the following chemicals: 2-bromoethanol from Tosoh Corporation, lithium bis(trifluoromethanesulfonyl) imide (LiTFSI) from Sumitomo 3M Co. chloromethylstyrene (*meta*, *para*-isomer mixture) from Seimi Chemical Co. Ltd, ethylene glycol dimethacrylate (EGDM) from Mitsubishi Rayon Co. Ltd, 1,4-dibromobutane and 1,6-dibromohexane from Tokyo Kasei Kogyo Co. Ltd; 1-methylimidazole and 1-ethyl-3-methylimidazolium tetrafluoroborate (EMIBF4) from Sigma-Aldrich Corporation; dimethyl 2,2′-azobis(isobutyrate) (MAIB), sodium hydroxide, imidazole, methanol, acetonitrile, toluene, dimethylsulfoxide (DMSO), dichloromethane, and *N*,*N*-dimethylformamide (DMF) from Wako Pure Chemical Industries, Ltd; *N*,*N*′-dicyclohexylcarbodiimide, acrylic acid, diethylether, and 2-bromoethane from Kanto Kagaku. All reagents were used without further purification. EMIBF4 was dried under vacuum before use.

2.2. Synthesis

2.2.1. 1,4-Bis(imidazole-1-yl)butane

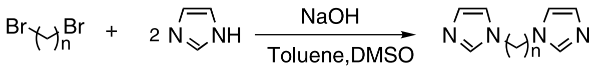
The synthesis procedure was carried out according to the literature [16].

^{1}H NMR(400 MHz, DMSO-d_6, ppm): δ 7.61 (s, 2H), 7.14 (d, 2H, $J=1.1$ Hz), 6.88 (d, 2H, $J=1.1$ Hz), 3.96 (t, 4H, $J=6.6$ Hz), 1.66–1.57 (m, 4H, $J=6.6$ Hz) (Scheme 1).

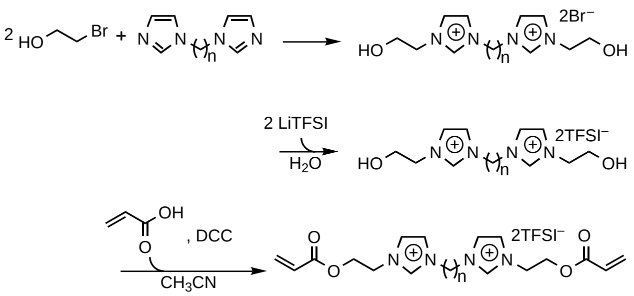
2.2.2. 1,6-Bis(imidazole-1-yl)hexane

Following the similar manner for the 1,4-bis(imidazole-1-yl)butane, imidazole (68.0 g, 1 mol) was dissolved in toluene (120 ml) and DMSO (120 ml) in a flask. After adding 50% aqueous solution of NaOH (80 g), water was removed by azeotropic distillation through a Dean-Stark condenser. DMSO (60 ml) was then added to the solution and 1,6-dibromohexane (122.0 g, 0.5 mol) was added in dropwise. Then the reaction mixture was stirred at 120 °C for 2 h. The deposit NaBr was thermally filtrated and solvent was removed under vacuum. The resulting crude solution was dissolved in dichloromethane (200 ml), and the residual NaBr was filtered off. After removal of the solvent, the 1,6-bis(imidazole-1-yl)hexane was obtained as a slightly yellowish solid (101.2 g, 93% yield).

^{1}H NMR(400 MHz, CDCl3, ppm): δ 7.44 (s, 2H), 7.05 (bs, 2H), 6.88 (s, 2H), 3.91 (t, 4H, $J=7.0$ Hz), 1.78–1.72 (m, 4H, $J=7.0$ Hz), 1.31–1.28 (m, 4H).



Scheme 1.



1-Cn (n = 4 or 6)

Scheme 2.

2.2.3. 1,4-Bis{3-[2-(acryloyloxy)ethyl]imidazolium-1-yl} butane bis[bis(trifluoromethane-sulfonyl)imide] (**1**-C4)

In a reactor, 1,4-bis(imidazole-1-yl)butane 5.71 g (30 mmol) and 2-bromoethanol (22.60 g, 181 mmol) were dissolved in DMF (10 ml) and the mixture was stirred at 50 °C for 3 days. The reaction mixture was reprecipitated in diethyl ether (200 ml, twice), to obtain imidazolium bromide as a yellowish solid (13.2 g, 100%). The bromide salt (6.60 g, 15 mmol) was dissolved in 150 ml of ion-exchange water, where an anion exchange reaction occurred after adding LiTFSI (8.7 g, 30 mmol). Washing the residual insoluble material with water produced TFSI salt as a slightly yellowish liquid (6.39 g, 51%). Dried TFSI salt and acrylic acid (5.6 g, 77.7 mmol) were dissolved in acetonitrile (80 ml) and then *N*, *N*′-dicyclohexylcarbodiimide (9.45 g, 45.8 mmol) was added as the mixture was cooled in an ice bath. After reaction at room temperature for 5 h under bubbling air, the solvent was removed, the residual urea was filtrated, and then the crude monomer was purified by washing with diethylether and dried under vacuum to give **1**-C4 (6.32 g, 6.7 mmol, 88% yield from TFSI salt) as a slightly yellowish liquid.

^{1}H NMR(400 MHz, DMSO-d_6, ppm): δ 9.21 (s, 2H), 7.84–7.83 (m, 2H), 7.77 (m, 2H), 6.33 (dd, 2H, $J=1.5$, 17.2 Hz), 6.15 (dd, 2H, $J=10.3$, 17.2 Hz), 5.98 (dd, 2H, $J=1.5$, 10.3 Hz), 4.52–4.48 (m, 8H), 4.27–4.22 (m, 4H), 1.77–1.76 (m, 4H) (Scheme 2).

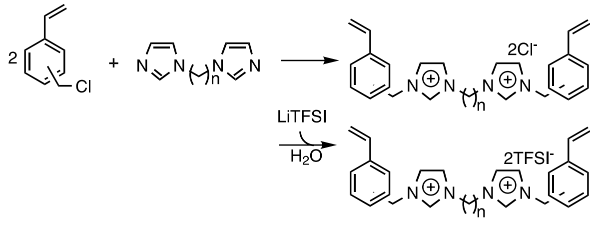
2.2.4. 1,6-Bis{3-[2-(acryloyloxy)ethyl]imidazolium-1-yl} hexane bis[bis(trifluoromethane-sulfonyl)imide] (**1**-C6)

The synthesis procedure followed the method of **1**-C4. The objective product **1**-C6 was obtained as a slightly yellowish liquid (68% yield).

^{1}H NMR(400 MHz, DMSO-d_6, ppm): δ 9.21 (s, 2H), 7.82 (m, 2H), 7.78 (m, 2H), 6.33 (dd, 2H, $J=1.5$, 17.4 Hz), 6.15 (dd, 2H, $J=10.3$, 17.4 Hz), 5.98 (dd, 2H, $J=1.5$, 10.3 Hz), 4.52–4.44 (m, 8H), 4.18–4.12 (m, 4H), 1.77–1.76 (m, 4H). 1.25–1.23 (m, 4H) (Scheme 3).

2.2.5. 1,4-Bis[3-(vinylbenzyl)imidazolium-1-yl]butane bis[-bis(trifluoromethanesulfonyl)-imide], isomer mixture (**2**-C4)

In a reactor, 1,4-bis(imidazole-1-yl)butane 1.904 g (10.0 mmol) and chloromethylstyrene 9.2 g (60 mmol) were dissolved in DMF (10 ml) and the mixture was stirred at 50 °C for 2 days. The reaction mixture was reprecipitated into



2-Cn (n = 4 or 6)

Scheme 3.

diethylether (200 ml, twice), to obtain imidazolium chloride as a slightly yellowish liquid. The chloride salt was dissolved in 150 ml of ion-exchange water, where an anion exchange reaction occurred after adding LiTFSI (5.9 g, 20.6 mmol). Washing the residual insoluble material with water and drying under vacuum produced monomer **2**-C4 as a slightly yellowish liquid (8.49 g, 86%).

^{1}H NMR(400 MHz, DMSO-d_6, ppm): δ 9.26, 9.24 (s, 2H), 7.85–7.78 (m, 4H), 7.54–7.29 (m, 8H), 6.79–6.71 (m, 2H), 5.90–5.85 (m, 2H), 5.41, 5.40 (s, 2H), 5.35–5.30 (m, 2H), 4.21 (m, 4H), 1.79 (m, 4H).

2.2.6. 1,6-Bis[3-(vinylbenzyl)imidazolium-1-yl]hexane bis[-bis(trifluoromethanesulfonyl)-imide], isomer mixture (2-C6)

Following the similar procedure as that described for **2**-C4, **2**-C6 was obtained as a slightly yellowish liquid (7.81 g, 77%).

^{1}H NMR(500 MHz, DMSO-d_6, ppm): δ 9.22, 9.20 (s, 2H), 7.78–7.73 (m, 4H), 7.48–7.26 (m, 8H), 6.71–6.66 (m, 2H), 5.83–5.79 (m, 2H), 5.36, 5.35 (s, 2H), 5.27–5.23 (m, 2H), 4.02–4.16 (m, 4H), 1.73 (m, 4H), 1.21 (m, 4H).

2.2.7. 1-[2-(Acryloyloxy)ethyl]-3-ethylimidazolium bis(trifluoromethanesulfonyl)imide (**3**)

For the general preparation method of ionic liquid monomers, we followed the procedure in our previous papers [14,17]. 2-Bromoethyl acrylate 17.9 g (100 mmol) was stirred with *N*-ethylimidazole 14.4 g (150 mmol) at room temperature for 24 h. The mixture was reprecipitated into diethylether (300 ml, twice) to give imidazolium bromide as a yellowish solid. The bromide salt was dissolved in 200 ml of ion-exchange water, in which an anion exchange reaction occurred after adding LiTFSI (22.1 g, 77.0 mmol). Washing the residual insoluble material with water and drying under vacuum produced **3** as a slightly yellowish liquid (24.39 g, 51%).

^{1}H NMR(400 MHz, DMSO-d_6, ppm): δ 9.22 (s, 1H), 7.81–7.80 (m, 2H), 6.35 (dd, 1H, $J=1.5$, 17.3 Hz), 6.18 (dd, 1H, $J=10.4$, 17.3 Hz), 5.99 (dd, 1H, $J=1.5$, 10.4 Hz), 4.53–4.47 (m, 4H), 4.22 (q, 2H, $J=7.3$ Hz), 1.42 (t, 3H, $J=7.3$ Hz).

2.2.8. 1-Ethyl-3-methylimidazolium bis(trifluoromethanesulfonyl)imide (EMITFSI)

The synthesis followed the procedure in reference [1(b)].

^{1}H NMR(400 MHz, DMSO-d_6, ppm): δ 9.09 (s, 1H), 7.77–7.68 (m, 2H), 4.19 (q, 2H, $J=7.3$ Hz), 3.84 (s, 3H), 1.42 (t, 3H, $J=7.3$ Hz) (Chart 1).

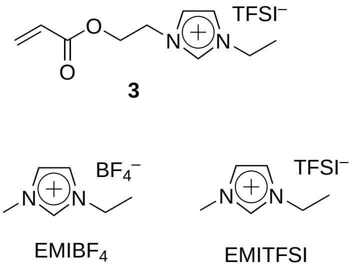


Chart 1.

2.3. Polymerization of ionic liquid monomers

Ionic liquid monomer (**3**) was mixed with MAIB (2 mol% to the vinyl unit) and suitable amount of cross-linker in the bulk, and then the mixture was put between two glass plates 0.5 mm apart for polymers and 1.3 mm for monomers. They were polymerized in the bulk under nitrogen atmosphere at 70 °C for 8 h. The resulting polymers were dried under vacuum at 60 °C for 24 h. The sample was further dried at 80 °C for 6 h before measurement. FT-IR measurement revealed that there was no monomeric species remained in the matrix after polymerization. Details see Methods.

2.4. Polymerization of ionic liquid monomers in the presence of imidazolium salts

Polymerizations in imidazolium ILs were performed in a similar manner as that described above. Designated amount of cross-linker and imidazolium ILs were added to the mixture of monomer and MAIB to produce a salt-containing-polymers.

2.5. Methods

The structure of these ionic liquid monomers were confirmed by ^{1}H NMR spectroscopy (JEOL α-500 or ECP 400 NMR spectrometer). After polymerization, absence of monomer is confirmed by FT-IR measurement, that is, peak for carbon–carbon double bond disappeared completely in the ionic liquid polymers. And the obtained polymer was used without further purification. Because when we apply this network polymer consist of the compounds, which have ionic liquid moiety to devises, it is useful that we can use the polymer compound obtained in a cell. Cross-linked polymer could not be determined its molecular weight by usual method, polymerization degree of these compounds is not clear. But film or rubber-like properties clearly indicated that monomer change to polymers. The ionic conductivity of the derived monomers and polymers were measured with the complex-impedance method using an impedance analyzer (Solartron gain phase analyzer, Model 1260, Schlumberger). For the dynamic ionic conductivity measurements, we used a system that was developed in our laboratory [18], and the measurements were carried out in a glove box filled with dry nitrogen gas in the temperature range of 10–60 °C. The DSC (differential scanning calorimetry) measurements were using either DSC-6200 (Seiko Instruments Inc.) or Q-100 (TA Instruments) at a heating rate of 10 °C min^{-1}. The glass transition temperature (T_g) was measured using the onset method. To determine the thermal stability of the prepared samples, we used Q-500 (TA Instruments) in the temperature range of 25–500 °C at a heating rate of 10 °C min^{-1}. The T_d of the samples was determined by onset and 10% weight-loss temperature.

3. Results and discussion

3.1. Homopolymerization of new cross-linking monomer containing ionic liquid structure

The summary of the thermal gravimetric analysis of the polymerized cross-linker monomers is listed in Table 1. The T_d in this table was determined from onset and 10% weight-loss temperature. All systems were thermally stable and their T_d was found at above 400 °C. This is attributable to the ionic liquid moiety. This suggested there is no fear of thermal degradation starting from the cross-links. However there are a few papers on the thermal properties of ionic liquid polymers, there is no data on the thermal properties of the polymerized ionic liquids having multi-polymerizable groups.

These cross-linkers produced matrix polymers of high T_d, and some of ionic conductivity was expected because of polymerized ionic liquids with free anions. However, the ionic conductivity of these polymers was very low. This is because of both the lack of successive ion conduction path with high segmental motion. In order to increase ionic conductivity, the domain with low T_g was introduced to polymer of **1**-C4.

3.2. Copolymerization of ionic liquid monomer and cross-linking monomer

Vinyl monomer **3** was polymerized in the presence of the cross-linking monomer, **1**-C4. Fig. 1 represents the Arrhenius plots of ionic conductivity for the obtained network polymers. These network polymers showed good ionic conductivity. The homopolymer of **3** was not cross-linked and, therefore, showed good ionic conductivity as shown in Fig. 1, closed circles. The TFSI anions acted as mobile ions because all of imidazolium cations were fixed on the main chain. The ionic conductivity decreased with increasing **1**-C4 content. In spite of small amount of **1**-C4, the cross-linked polymer certainly showed lower ionic conductivity. When ionic liquid monomer was polymerized in the presence of polyether-type cross-linker, little decrease in the ionic conductivity was found [15]. This is one of advantages of the cross-linker with polyether chains. However, the system containing polyether chains has poor thermal stability. Against this, the present system showed a little decrease in the ionic conductivity in the case of small amount of cross-linker **1**-C4 was added. The decrease in the ionic conductivity was reasonably monotonic until 5 mol%, the conductivity of the cross-linked polymer with 10 mol% **1**-C4 was not low as expected from the data for 5 mol% one.

Table 1
Thermal gravimetric analysis data of polymerized cross-linking monomers

Compound	T_d (°C)	
	Onset	10% weight-loss
1-C4	415	412
1-C6	415	405
2-C4	421	419
2-C6	432	429

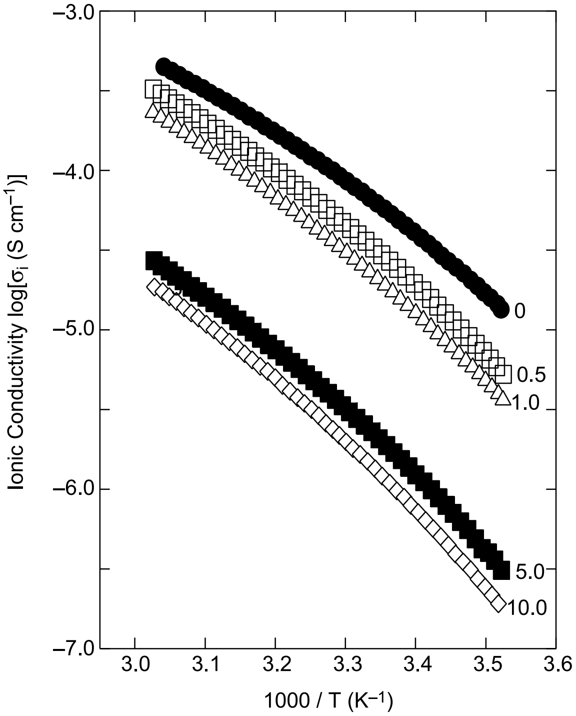


Fig. 1. Temperature dependence of the ionic conductivity for cross-linked copolymers of **3** and **1**-C4. Numbers in the figure mean the amount of **1**-C4 (mol%).

As expected from excellent thermal stability of **1**-C4, these cross-linked polymers certainly showed better properties than ordinary polyether-containing systems. Results of the thermal gravimetric analysis of the network polymers are shown in Fig. 2. To clarify the differences of cross-linker with or without IL moiety, EGDM was used as cross-linker. By compare the thermogram for homopolymer of **3**, there is no decline when the monomer **3** was copolymerized with 10 mol% **1**-C4. Against this, certain drop of Td was found when **3** was copolymerized with the same amount of EGDM. The thermal stability was found to be suppressed when mono-functional ionic liquid monomer **3** was copolymerized with EGDM. It is clear that the ionic liquid type cross-linker did not affect the T_d because it has sufficiently high T_d (>400 °C). Thus the introduction of ionic liquid moiety to both acrylate monomers and cross-linkers elevated the thermal stability of the corresponding polymer networks.

3.3. The ionogel containing ethylimidazolium salts

To improve the ionic conductivity of the network polymers, the effect of ethylmethylimidazolium (EMI) salts was

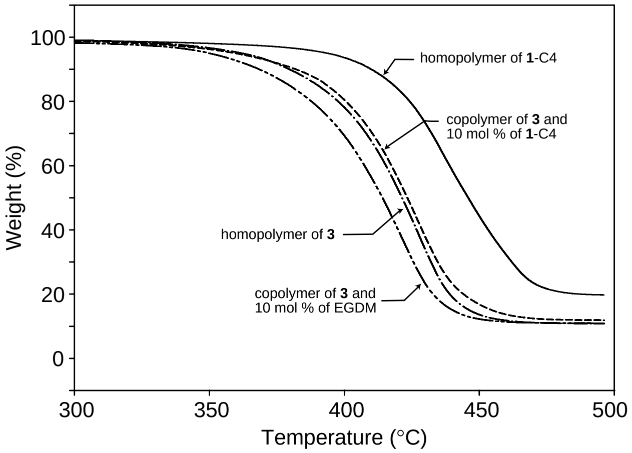


Fig. 2. TG curves for polymers. The heating rate was 10 °C min^{-1}. Solid line, homopolymer of **1**-C4; short-dash, copolymer of **3** and 10 mol% of **1**-C4; dash-dot, homopolymer of **3**; broken double dot, copolymer of **3** and 10 mol% of EGDM.

investigated. Fig. 3 shows the correlation between ionic conductivity and salt concentration for a series of the obtained ionogels.

The ionic conductivity of the ionogel increased with an increasing amount of EMIBF4. The polymer without EMIBF4 was anion conductor, because all the cations were fixed on the main chain. After adding EMIBF4, it became bi-ion conductor. And considerable increase in the ionic conductivity was found. The ionic conductivity exceeded that of the monomer (3.63×10^{-3} S cm^{-1}, at 50 °C) when 200 mol% EMIBF4 was added.

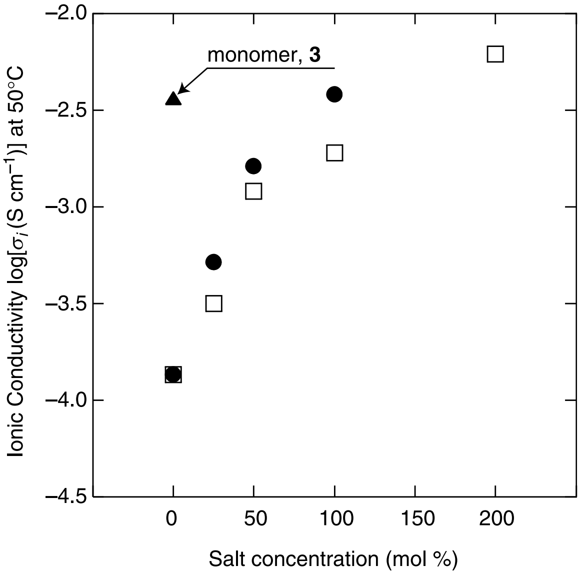


Fig. 3. Effect of added salt concentration on the ionic conductivity of the polymerized **3** in the presence of cross-linker **1**-C4 (1 mol%). □; EMIBF4, ●; EMITFSI The ionic conductivity of monomer **3** was also shown as reference (closed triangle).

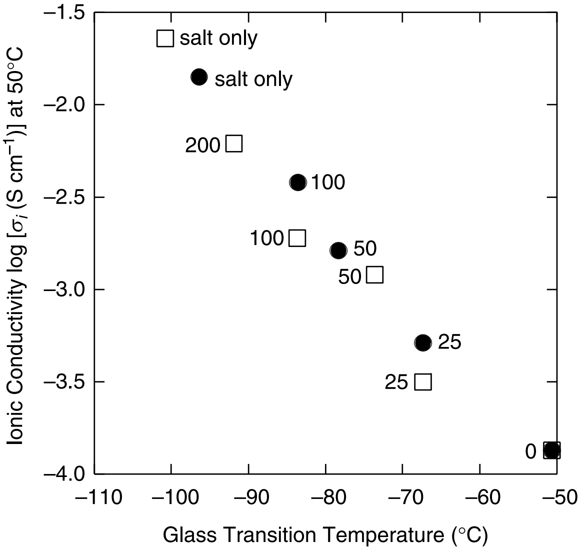


Fig. 4. Relationship between T_g and ionic conductivity at 50 °C for the polymerized 3 in the presence of cross-linker **1**-C4 (1 mol%) containing EMI salts (□; EMIBF4, ●; EMITFSI). The numbers in the figure are the salt concentration (mol%).

It is generally known that the ionic conductivity is the function of T_g when ion migration was coupled with the segmental motion of the matrix [19]. This discussion is effective for the ionogels where the ion conductive pathway was composed by the ionic liquid. In such a case, the ionic conductivity of the ionogels is the function of the T_g of the added ionic liquids. The relationship between T_g and ionic conductivity in these salt-added systems is shown in Fig. 4. When the amount of salt increased, T_g was lowered and accordingly the ionic conductivity increased. These results show that the addition of EMIBF4 increased the ionic conductivity due to both the increase of number of ions and the lowered T_g value. From this relation, it is clear that the added EMIBF4 is effective to construct successive ion conduction path because of very good affinity with the polymer matrix applied here.

The influence of the incorporation of the EMI salt for the polymerization reaction was not analyzed. The ionogel containing EMI may have less mechanical properties with increasing EMI content, because EMI is expected to play as a plasticizer in an ionogel. Since, the main purpose of this study was to improve the thermal stability of ion conductive polymers, the TGA measurement was carried out for **3** which was polymerized in the presence of a cross-linker and EMI salts. Fig. 5 shows their typical thermograms. The weight of the EMIBF4-containing polymer started to decrease at around 300 °C. On the other hand, the ionogel with 100 mol% of EMITFSI showed good compatibility. This EMITFSI ionogel had a high decomposition temperature of about 400 °C due to excellent thermal stability of TFSI anions. This also should be noted here that the selection of anions for additive salts is quite important to design thermally stable polymer matrix. It is without saying that the matrix should be stable enough, otherwise the mixture gradually degraded even after addition of these stable additives. Talking these into account, the ionic liquid-based cross-linkers are quite useful for designing thermally stable polymer network. Thus, we confirmed that a highly thermally-stable ionogel was obtained by adding EMITFSI to these polymers.

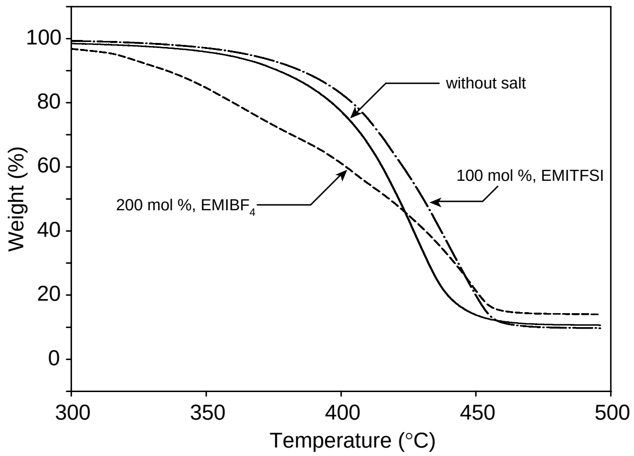


Fig. 5. TG curves for polymerized **3** in the presence of cross-linker **1**-C4 (1 mol%) and EMI salts. The heating rate was 10 °C min^{-1}.

4. Conclusions

Synthesis and polymerization of new cross-linking monomers based on the ionic liquid were investigated to obtain thermally stable polymers with high ionic conductivity. Homopolymers of new cross-linking monomers showed good thermal stability above 400 °C. These ionic liquid-based cross-linkers are effective to design thermally stable network polymer structure. Copolymerization of mono acrylic-type ionic liquid monomer with a new cross-linker (**1**-C4, 1 mol%) led to a polymer of high ionic conductivity: 1.36×10^{-4} S cm^{-1} (at 50 °C). This network polymer also had good thermal stability (Tdonset; 394 °C). Copolymerization in ethylimidazolium-type ILs induced the increase of ionic conductivity in comparison with that of monomers. A transparent film was obtained by adding 50 mol% of EMITFSI. The thermal decomposition temperature was kept to be about 400 °C when EMI salt was added to the network polymer. Also, a novel ion conductive matrix was prepared by copolymerization of the mono-acrylate monomer with the cross-linking monomer in the presence of ethylimidazolium ionic liquid. Thus we succeeded in obtaining films with high ionic conductivity and very high thermal stability.

Acknowledgement

The present study was supported by a Grant-in-aid for Scientific Research from the Ministry of Education, Culture, Sports, Science and Technology (#14205136 and #17205020). The present study was carried out under the 21st Century COE program, Future Nano Materials. We thank the Tosoh Corporation for giving us the 2-bromoethanol, Sumitomo 3M Co. for lithium bis(trifluoromethanesulfonyl)imide, Seimi Chemical Co. Ltd, for chloromethylstyrene, and Mitsubishi Rayon Co. Ltd for ethyleneglycol dimethacrylate.

References

[1] (a) Wilkes JS, Zaworotko MJ. J Chem Soc Chem Commun 1992;965.
  (b) Bonhôte P, Dias A-P, Papageorgiou N, Kalyanasundaram K, Grätzel M. Inorg Chem 1996;35:1168.
  (c) Wasserscheid P, Welton T, editors. Ionic liquids in synthesis. Weinheim: Wiley-VCH; 2003.
[2] Welton T. Chem Rev 1999;99:2071.
[3] (a) Carlin RT, De Long HC, Fuller J, Trulove PC. J Electrochem Soc 1994;141:L73.
  (b) Koch VR, Nanjundiah C, Appeetecchi GB, Scrosati B. J Electrochem Soc 1995;142:L116.
  (c) Yoshio M, Mukai T, Kanie K, Yoshizawa M, Ohno H, Kato T. Chem Lett 2002;320.
  (d) Hayashi A, Yoshizawa M, Angell CA, Mizuno F, Minami T, Tatsumisago M. Electrochem Solid-State Lett 2003;6:E19.
  (e) Ogihara W, Yoshizawa M, Ohno H. Chem Lett 2002;880.
[4] (a) Nakagawa H, Izuchi S, Kuwana K, Aihara Y. J Electrochem Soc 2003;150:A695.
  (b) Sun J, MacFarlane DR, Forsyth M. Solid State Ionics 2002;147:333.
  (c) Forsyth M, Sun J, Zhou F, MacFarlane DR. Electrochim Acta 2003;48:2129.
  (d) Ohno H, Yoshizawa M, Ogihara W. Electrochim Acta 2003;48:2079.
[5] (a) Doyle M, Choi SK, Proulx G. J Electrochem Soc 2000;147:34.
  (b) Sun J, Jordan LR, Forsyth M, Macfarlane DR. Electrochim Acta 2001;46:1703.
  (c) Sun J, MacFarlene DR, Forsyth M. Electrochim Acta 2003;48:1971.
[6] (a) Kubo W, Makimoto Y, Kitamura T, Wada Y, Yanagida S. Chem Lett 2002;948.
  (b) Wang P, Zakeeruddin SM, Exnar I, Grätzel M. Chem Commun 2002; 2972.
[7] (a) Lewandowski A, Swiderska A. Solid State Ionics 2003;161:243.
  (b) Ue M, Yakeda M, Toriumi A, Kominato A, Hgiwara R, Ito Y. J Electrochem Soc 2003;150:A499.
  (c) Stenger-Smith JD, Webber CK, Anderson N, Chafin AP, Zong K, Reynolds R. J Electrochem Soc 2002;149:A973.
[8] (a) Lu W, Fadeev AG, Qi B, Smela E, Mattes BR, Ding J, et al. Science 2002;297:983.
  (b) Zhou D, Spinks GM, Wallace GG, Tiyaboonchaiya C, MacFarlane DR, Forsyth M, et al. Electrochim Acta 2003;48:2355.
[9] Baranyai KJ, Deacon GB, MacFarlane DR, Pringle JM, Scott JL. Aust J Chem 2004;57:145.
[10] Hirao M, Ito K, Ohno H. Electrochim Acta 2000;45:1291.
[11] Ohno H, Ito K. Chem Lett 1998;751.
[12] Yoshizawa M, Ogihara W, Ohno H. Polym Adv Technol 2002;13:589.
[13] Yoshizawa M, Hirao M, Ito-Akita K, Ohno H. J Mater Chem 2001;11: 1057.
[14] Yoshizawa M, Ohno H. Electrochim Acta 2001;46:1723.
[15] Washiro S, Yoshizawa M, Nakajima H, Ohno H. Polymer 2004;45:1577.
[16] So Y-H. Macromolecules 1992;25:516.
[17] Ito K, Nakai Y, Omori M, Ohno H. Polymer Prep Jpn 1997;46:510.
[18] Ohno H, Inoue Y, Wang P. Solid State Ionics 1993;62:257.
[19] Ohno H, Yoshizawa M, Ogihara W. Electrochim Acta 2004;50:255.