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Highly Conductive Ionic-Liquid Gels Prepared with Orthogonal Double Networks of a Low-Molecular-Weight Gelator and Cross-Linked Polymer

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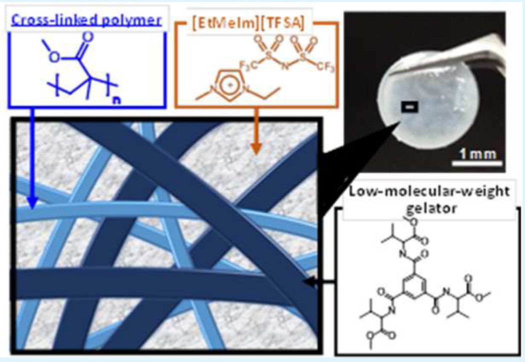
Supporting Information



ABSTRACT: We prepared a heterogeneous double-network (DN) ionogel containing a low-molecular-weight gelator network and a polymer network that can exhibit high ionic conductivity and high mechanical strength. An imidazolium-based ionic liquid was first gelated by the molecular self-assembly of a low-molecular-weight gelator (benzenetricarboxamide derivative), and methyl methacrylate was polymerized with a cross-linker to form a cross-linked poly(methyl methacrylate) (PMMA) network within the ionogel. Microscopic observation and calorimetric measurement revealed that the fibrous network of the low-molecular-weight gelator was maintained in the DN ionogel. The PMMA network strengthened the ionogel of the low-molecular-weight gelator and allowed us to handle the ionogel using tweezers. The orthogonal DNs produced ionogels with a broad range of storage elastic moduli. DN ionogels with low PMMA concentrations exhibited high ionic conductivity that was comparable to that of a neat ionic liquid. The present study demonstrates that the ionic conductivities of the DN and single-network, low-molecular-weight gelator or polymer ionogels strongly depended on their storage elastic moduli.

KEYWORDS: *conducting materials, gels, ionic liquids, polymers, molecular self-assembly*

INTRODUCTION

Over the past two decades, ionic liquids have received considerable attention due to their high ionic conductivity, wide electrochemical window, high chemical and thermal stabilities, inflammability, and tunable properties.[1] Owing to these attractive characteristics, ionic liquids have been studied as electrolytes in fuel cells, solar cells, batteries, and sensors.[2−6] There has been, especially, a considerable demand for the preparation of highly conductive, flexible, and transparent solid polyelectrolytes with diverse configurations, which can prevent battery leaks on impact. There are several approaches to immobilize ionic liquids: gelation, impregnation of porous substrates with ionic liquids,[7,8] and polymerization of ionic liquids.[9−12] Among them, gelation is a facile and effective approach to immobilizing ionic liquids to avoid the loss of their characteristics. To date, several strategies have been proposed to gel ionic liquids: gelation by cross-linked polymer,[13,14] by supramolecularly cross-linked polymer,[15] by interconnected network of inorganic oxides,[16−18] and by molecular self-assembly of a low-molecular-weight gelator.[19−24]

Susan et al. reported the in situ polymerization of methyl methacrylate (MMA) in ionic liquids and succeeded in the preparation of cross-linked poly(methyl methacrylate) (PMMA) ionogel.[25] They also studied the ionic conductivity of the ionogel in detail and revealed that the conductivity decreased with the increase of the PMMA concentration. The conductive ionogels prepared with cross-linked block copolymers with high mechanical strength were also reported by the Lodge and Watkins groups.[26,27] Neouze et al. studied the preparation of ionogels using in situ sol−gel reaction with a high content of silica.[18] Hanabusa et al. succeeded in the gelation of ionic liquids using low-molecular-weight gelators at low concentrations (∼1 wt %), which did not reduce their intrinsic ionic conductivities.[20] Yang et al. reported that their low-molecular-weight gelators had negligible effects on the ionic conductivity and on the electrochemical window of ionic liquids.[28,29] Schubert et al. found the thermoreversible gelation of an amphiphilic quaternary ammonium-type ionic liquid/water mixture, which exhibited high conductivity probably due to the controllable water content and the additional inorganic salts.[30−32] We also reported the gelation of ionic liquids using novel low-molecular-weight gelators and revealed that the gelation did not affect the ionic conductivities of ionic liquids.[23,24] Yanagida et al. demonstrated the feasibility of ionogels to the solid-state dye-sensitized solar cells using a low-molecular-weight gelator.[33] However, gelation by low-molec-

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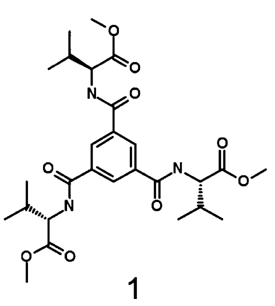
ular-weight gelators in most cases produces gels with remarkably low mechanical strengths compared to polymer-based gels, leading to solation or bleeding.[34] Because the portable batteries and electronic devices have high risks of collision and dropping, the materials that constitute the batteries and devices should have resistance to physical impacts.

To overcome the brittleness of ionogels prepared with low-molecular-weight gelators and also to maintain the intrinsic ionic conductivity, here we prepare a heterogeneous double-network (DN) ionogel containing a self-assembled network of low-molecular-weight gelators and a cross-linked polymer. The concept of a double-network gel was first proposed by Gong et al. to achieve a high mechanical strength in hydrogels, by synthesizing two independent polymer networks that contributed cooperatively to the relaxation and dissipation of applied stress, leading to supertough hydrogels.[35−37] On the other hand, there are several studies on polymerizing organogels with low-molecular-weight gelators that employed monomer solutions as solvents and produced a solid polymer (rather than a gel), embedded with self-assembled networks of low-molecular-weight gelators.[38−40] To our knowledge, there is no paper describing a heterogeneous double-network gel containing a self-assembled network of low-molecular-weight gelators and a cross-linked polymer network. In the present study, we prepare conductive DN ionogels using the molecular self-assembly of low-molecular-weight gelators (first network) and a cross-linked polymer network (second network) and reveal the correlations between mechanical properties and ionic conductivity. The conductive ionogels with high mechanical strength will extend the practical application of ionogels to solar cells, batteries, capacitors, etc.

RESULTS AND DISCUSSION

DN Gels of Ionic Liquids. In our previous work, we developed novel low-molecular-weight gelators with simple molecular structures as ionic liquids.[24] Gelator molecules contained 1,3,5-benzenetricarboxylic acid and amino acid methyl esters (Chart 1). Intermolecular hydrogen bonding between amide groups and π−π interactions are important in the fibrous self-assembly of gelator molecules in ionic liquids as indicated by papers describing the assembly of benzenetricarboxamide derivatives in organic solvents.[41−45] We prepared DN ionogels containing two independent networks, a molecular self-assembled network of a low-molecular-weight gelator (first network), and a cross-linked PMMA (second network). The DN ionogel was prepared in two steps. First, an ionogel of a low-molecular-weight gelator was prepared by heating [EtMeIm][TFSA] (Figure S1) containing gelator **1** (1.0 wt %) to ~150 °C and subsequently cooling to room temperature. Second, the ionogel was immersed in [EtMeIm][TFSA] containing 20 wt % MMA, ethylene glycol dimethacrylate (EGDMA, 2.0 mol % of MMA) and 2,2′-azobis(2-methylpropionitrile). After immersion for 24 h, the supernatant was removed, and the polymerization of MMA and EGDMA was performed at 60 °C for 24 h. Because the T_{gel} values of the ionogels with gelators **1** and **2** were 82 and 90 °C, respectively,[24] the polymerization proceeded in the gel state. The polymerization proceeded without apparent macroscopic phase separation. Hereafter, the DN ionogels are described as DN ionogel(x−y), where x and y indicate the weight percentages of a low-molecular-weight gelator and PMMA, respectively.

Chart 1. Molecular Structures of the Low-Molecular-Weight Gelators 1 and 2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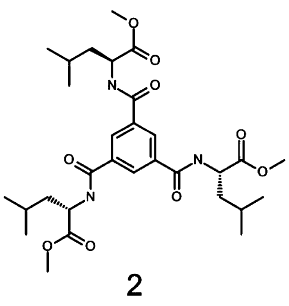
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As shown in Figure 1a,b, DN ionogel(1−9) was opaque and similar in appearance to the ionogels prepared with low-molecular-weight gelators (Figure S2a). The DN ionogels were robust and elastic, which allowed us to handle the ionogel using tweezers, whereas ionogels prepared only with low-molecular-weight gelators were mechanically weak and easily deformed. The DN ionogel was stable without bleeding of an ionic liquid for >3 months. When the polymer network was prepared in the ionic liquid before the self-assembly of the low-molecular-weight gelator, the DN ionogel was not obtained. The dense polymer network prevented the low-molecular-weight gelator from diffusing into the PMMA ionogel.

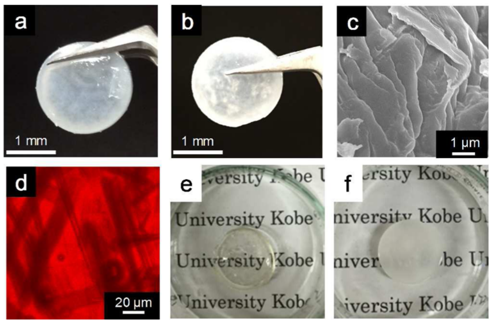

Figure 1. DN ionogel(1−9) prepared with gelators **1** (a) and **2** (b). (c) FE-SEM image of DN ionogel(1−9) prepared with [EtMeIm][TFSA] and gelator **1**. (d) CLSM image of DN ionogel(1−9) stained by 30 μM Rhodamine B. (e) DN ionogel(1−9) heated at 120 °C. (f) DN ionogel(1−9) cooled to room temperature after heating to 120 °C.

The DN ionogel structure was observed using FE-SEM and confocal laser scanning microscopy (CLSM) (Figure 1c,d). The FE-SEM image of the DN ionogel shows a creased nonfibrous structure, which was similar to that of the PMMA ionogel (Figure S2b). This result indicates the dense network of cross-linked PMMA in the DN ionogel, which is often observed in polymer gels.

Low-molecular-weight gelators produce entangled nanofibers or nanosheets via molecular self-assembly and solidify into a gel. CLSM can provide in situ cross-sectional observation of gels prepared with low-molecular-weight gelators,[23,45,46] whereas preparation for FE-SEM observation requires solvent exchange, freeze-drying, and metal-sputtering and provides only surface morphology. DN ionogel(1−9) prepared with gelator **1** was stained with 30 μM Rhodamine B for CLSM observation. Figure 1d shows that Rhodamine B was found between fibers, which was consistent with our previous results with an ionogel of gelator **1**.[24] Rhodamine B is highly soluble in

[EtMeIm][TFSA] (>1 g/L) but immiscible in gelator **1**, resulting in the exclusion from the self-assembled network of gelator **1**. This result suggests that the DN ionogel maintained the fibrous structure derived from the molecular self-assembly of gelator **1** even after the polymerization of MMA and EGDMA in the ionogel.

Although the ionogel prepared only with PMMA was transparent (Figure S2c), the DN ionogel was opaque. The ionogel of gelator **1** was also opaque because of the submicrometer fibers formed by the self-assembly of gelator molecules. The opaqueness of the DN ionogel implies the presence of gelator **1** fibers. Heating to 120 °C made the DN ionogel transparent (Figure 1e), and subsequent cooling to room temperature reverted it to opaque (Figure 1f). This reversible change in appearance is because gelator **1** reassembles within the PMMA ionogel.

The benzenetricarboxamide-based gelators we previously reported can solidify various ionic liquids into gels.[24] We have examined the preparation of DN ionogels using other ionic liquids ([BuMeIm][TFSA] and [BuPy][TFSA]). Figure S3 shows the photos of the DN ionogels of [BuMeIm][TFSA] and [BuPy][TFSA]. These ionic liquids can also be applied as DN ionogels, indicating their versatility.

DSC Thermal Analysis of DN Ionogel. The DSC measurement was performed on DN ionogel(1−7) prepared with gelator **2**. An endothermic peak was observed at about 90 °C (Figure 2), which corresponds to a gel−sol transition of the ionogel of gelator **2**.[24] Although facile thermoreversible gel−sol transitions are a notable feature of low-molecular-weight gelators, the DN ionogel did not exhibit thermoreversible gel−sol transition between 0 and 200 °C because of the covalently cross-linked PMMA network within the DN ionogel. The endotherm observed at about 90 °C demonstrates the presence of self-assembled gelator **2** in the DN ionogel and negligible interaction between self-assembled gelator **2** and a PMMA network, which agrees with the results in Figure 1e,f.

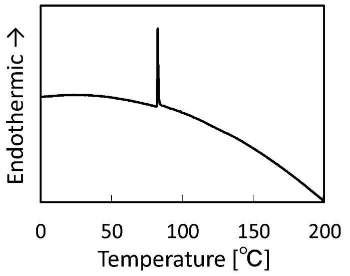

Figure 2. DSC thermogram for DN ionogel(1−7) at a heating rate of 10 °C/min. DN ionogel(1−7) was prepared with [EtMeIm][TFSA] and gelator **2**.

Characterization of MMA Polymerization in the DN Ionogel. To evaluate MMA polymerization in an ionogel, polymerization (9 wt % MMA) was carried out in the absence of cross-linker EGDMA in an [EtMeIm][TFSA] ionogel of 1 wt % gelator **1**. The gel permeation chromatography analysis of PMMA revealed M_n of 1.9 × 10^5 g/mol and M_w/M_n of 1.2. In the absence of gelator **1** and EGDMA, the polymerization of 9 wt % MMA in the ionic liquid resulted in M_n of 1.6 × 10^5 g/mol and M_w/M_n of 1.6, which were similar to those in the ionogel of 1 wt % gelator **1**. These results demonstrate that the free radical polymerization of MMA can proceed in the ionogel of a low-molecular-weight gelator.

A DN ionogel was prepared by immersing an ionogel of a low-molecular-weight gelator into an MMA solution and subsequently polymerizing MMA within the low-molecular-weight gelator network. Because the MMA concentration in the ionogel was not equal to that in solution, we estimated the concentrations of PMMA in the ionogel of gelator **1** after immersion and after polymerization. The MMA concentration was varied to estimate the PMMA content (Table 1). For example, when the initial MMA concentration was 20 wt %, the resulting PMMA concentration within the ionogel was 9.0 wt % (Table 1), implying that approximately half of the MMA polymerized in the ionogel. These results also indicate the infiltration of MMA and the successful PMMA polymerization in the ionogel of gelator **1**. Table 1 also shows the sample naming scheme of DN ionogel(*x*−*y*) corresponding to a DN ionogel prepared with *x* wt % gelator **1** and *y* wt % PMMA in the present study.

Table 1. PMMA Content in the Ionogel Prepared with [EtMeIm][TFSA] and 1 wt % Gelator 1

initial MMA concn (wt %) used	actual PMMA concn (wt %) in the resulting ionogel	DN ionogel name
10	7	DN ionogel(1−7)
20	9	DN ionogel(1−9)
30	13	DN ionogel(1−13)
40	16	DN ionogel(1−16)
50	26	DN ionogel(1−26)

Rheological Measurements of DN Ionogels. To evaluate the viscoelastic properties of the DN ionogels, dynamic frequency rheological measurements were carried out. The storage elastic modulus, *G*′, increased with increasing PMMA concentration (Figure 3a). Although the *G*′ value of the ionogel prepared only with gelator **1** was 4000 Pa, the presence of 9 wt % PMMA increased *G*′ by approximately 4-fold. The presence of 26 wt % PMMA increased *G*′ by 50-fold. Figure 3b

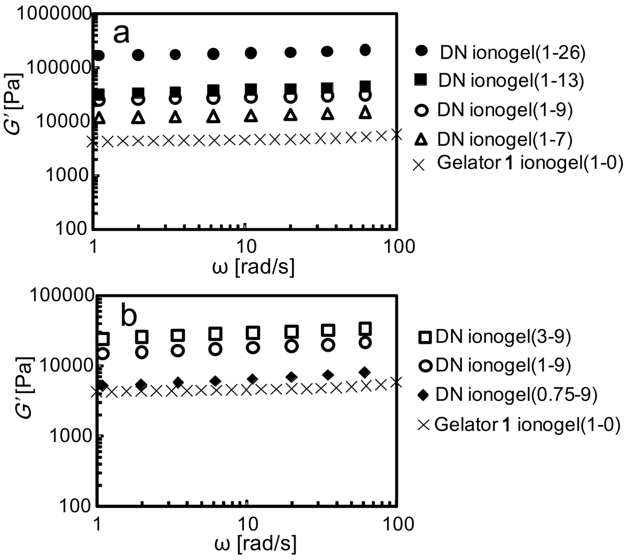

Figure 3. Viscoelastic properties of the DN ionogels. (a) Gelator **1** was used at 1.0 wt %, and the resulting PMMA content was varied from 7.0 to 26 wt %. (b) Gelator **1** was used at various concentrations from 0.75 to 3.0 wt %, and the resulting PMMA content was kept constant at 9.0 wt %. [EtMeIm][TFSA] was used as an ionic liquid.

shows the effect of the concentration of gelator **1**. Although PMMA at 9 wt % did not solidify [EtMeIm][TFSA] in the absence of gelator **1**, using only 0.75 wt % gelator **1** allowed gelation to occur, even though this concentration was lower than the critical gelation concentration. Gelator **1** at 3.0 wt % increased *G*′ 4-fold higher than that of DN ionogel(0.75−9). These results revealed that the storage elastic modulus of DN ionogel depended on both the concentrations of PMMA and gelator **1**. The presence of both the polymer and low-molecular-weight gelator networks effectively improved the mechanical strength of the ionogel. These results also indicate the synergetic effect of a DN gel even though the networks were independent of each other. The fragile gelator network absorbs crack energy through deformation, leading to increases in mechanical strength, whereas the PMMA network contributes to the elasticity. Gong et al. also reported an example of a DN hydrogel with a high mechanical strength and discussed the relaxation of locally applied stress as a combination of two independent gel networks.[35]

Compression Tests for the DN Ionogels. Figure 4 shows the stress−strain curves for the DN (a) and PMMA (b) ionogels tested under compression. For example, DN ionogel(1−9) and DN ionogel(1−13) failed at stresses of 0.95 and 0.47 MPa, respectively, whereas 20 wt % PMMA ionogel failed at 0.34 MPa. At a range of small strains (<0.4 mm/mm), the plots suggest that higher PMMA concentrations led to higher elastic moduli.

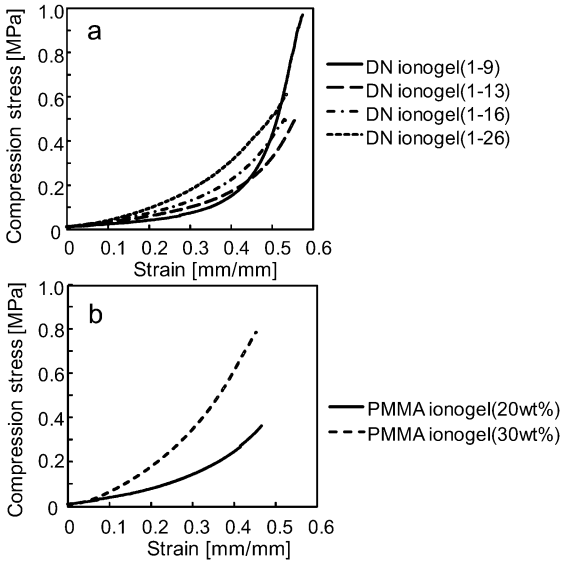

Figure 4. Stress−strain curves for [EtMeIm][TFSA] ionogels: (a) DN ionogels (1 wt % gelator **1**); (b) PMMA ionogels.

Figure 5 summarizes the effect of PMMA concentration on the fracture stress of DN and PMMA ionogels. Except for DN ionogel(1−9), the fracture stress of ionogels increased with the increase of PMMA concentration. DN ionogel(1−9) exhibited a remarkably high fracture stress despite the low PMMA concentration. The compression tests were repeated in triplicate for each sample and showed good reproducibility. These investigations revealed that the DN ionogel showed a high fracture stress with relatively low PMMA concentrations compared with those of PMMA ionogels.

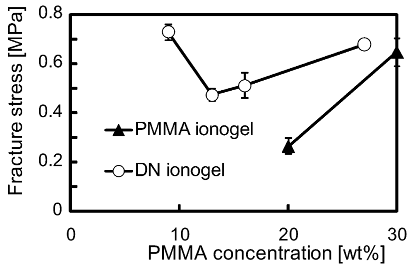

Figure 5. Effect of the PMMA concentration on the mechanical strength in the compression test of DN ionogels and PMMA ionogels. Gelator **1** was used at 1.0 wt % for DN ionogels. [EtMeIm][TFSA] was used as an ionic liquid.

Ionic Conductivity of DN Ionogels. Ionic liquids display specific electrochemical properties. Polymer-based gelation of ionic liquids has been reported to reduce the ionic conductivity of the resulting ionogels, probably because of the high polymer concentrations required for gelation.[14,25] However, ionogels prepared using low-molecular-weight gelators retain a high intrinsic conductivity of the corresponding ionic liquid.[20,23,24] The high conductivities of these ionogels may be due to relatively large voids between entangled fibers, which are related to the low gelator concentration required and facilitate the movement of ions. The ionic conductivities of DN ionogels prepared with gelator **1** and PMMA using [EtMeIm][TFSA] as a solvent were investigated. The DN ionogels were prepared with various MMA concentrations, and various *G*′ values were obtained. Figure 6a shows the effect of *G*′ on ionic conductivity.

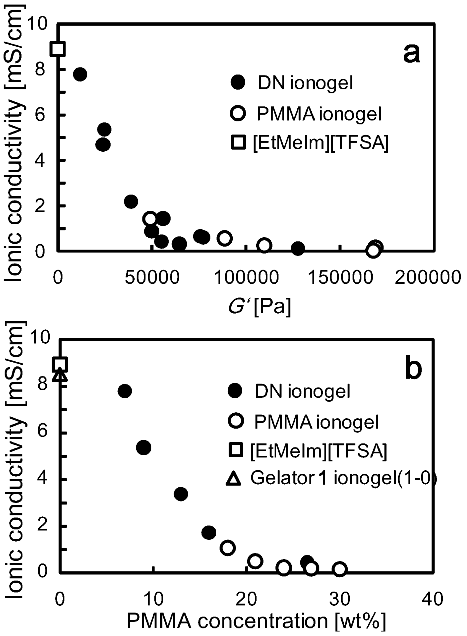

Figure 6. Ionic conductivities of the DN and PMMA ionogels: (a) effect of storage elastic modulus (*G*′); (b) effect of PMMA concentration. [EtMeIm][TFSA] was used as an ionic liquid.

The ionic conductivity of the DN ionogel decreased with increasing *G*′. Interestingly, PMMA ionogels showed *G*′ values similar to those of the DN ionogels, and they also exhibited similar ionic conductivities. DN ionogels with a *G*′ value <3000 Pa showed a relatively high ionic conductivity. Ionogels with a *G*′ value <3000 Pa cannot be prepared with PMMA alone.

Figure 6b summarizes the effect of MMA concentration on the ionic conductivities of DN ionogels and PMMA ionogels.

The ionic conductivity decreased with the PMMA concentration, indicating that the increase of the PMMA network inhibited the mobility of ions in the ionogels. The reducing conductivity in DN ionogels agreed well with those of PMMA ionogels. It should be noted that DN ionogel(1−9) exhibited 90% of the ionic conductivity of the neat ionic liquid. We previously reported that the concentration of gelator **1** does not affect the ionic conductivity of an ionogel prepared only with gelator **1**, and the ionogel of gelator **1** was too fragile to manipulate using tweezers.[24] The PMMA network can tightly immobilize more molecules of the ionic liquid compared with the fibrous self-assembly of a low-molecular-weight gelator alone. These results demonstrate that the ionic conductivity of a DN ionogel strongly depends on the polymer concentration, which is also related to the mechanical strength.

Cross-linking is another important factor for gelation. The effect of the cross-linker concentration on the ionic conductivity was investigated (Figure S4). The ionic conductivity decreased with the increase of the cross-linker concentration. This would be because the cross-linking largely affects the movement of solvent molecules. The low cross-linker concentration (0.5 mol %) can successfully prepare a DN ionogel, and its conductivity was comparable with that of the neat ionic liquid.

CONCLUSION

The present study is the first report of ionogels containing orthogonal DNs. One network is a fibrous molecular self-assembly of a low-molecular-weight gelator, and the other is a cross-linked polymer. The DN cooperatively facilitates the immobilization of ionic liquids and strengthens the ionogel. The DN ionogel provided a broad range of possible storage elastic moduli, which revealed a correlation between mechanical properties and the ionic conductivities of an ionogel. Among the DN ionogels prepared in the present study, the ionogel of a low-molecular-weight gelator (1 wt %) strengthened by a cross-linked polymer (9 wt %) exhibited the best performance (high ionic conductivity and high mechanical strength). This retained conductivity is because the ionic liquid is loosely immobilized at low concentrations of the low-molecular-weight gelator and cross-linked polymer. The present strategy of DN ionogels using self-assembly of a low-molecular-weight gelator and cross-linking a polymer is versatile and useful for a wide range of gels, which overcomes the brittleness of ionogels prepared only with low-molecular-weight gelators and coincidently avoids the loss of ionic conductivity in ionogels prepared only with cross-linked polymers. The ionogel maintains the functional properties of the host solvent and also benefits from the functional properties of a low-molecular-weight gelator and a cross-linked polymer that are similar to those of interpenetrated polymer hydrogels.[47]

EXPERIMENTAL SECTION

Materials. 1,3,5-Benzenetricarbonyl trichloride and methyl ester hydrochlorides of amino acids (L-valine, L-leucine) were purchased from Tokyo Chemical Industry (Tokyo, Japan). All solvents used in the syntheses were purchased from Wako Pure Chemical Industries (Osaka, Japan). [EtMeIm][TFSA], [BuMeIm][TFSA], and [BuPy][TFSA] were synthesized from [EtMeIm][Cl], [BuMeIm][Cl], [BuPy][Br], and [Li][TFSA] as reported previously.[21] Full names and chemical structures of the ionic liquids are provided in Figure S1. 2,2′-Azobis(2-methylpropionitrile) (AIBN) and methyl methacrylate (MMA) were purchased from Wako Pure Chemical Industries. Prior to polymerization, MMA was purified by an alumina column (Nacalai Tesque, Kyoto, Japan) to remove an inhibitor. Ethylene glycol dimethacrylate (EGDMA) was purchased from Tokyo Chemical Industry.

Synthesis. *Syntheses of Gelators 1 and 2.* Gelators **1** and **2** (Figure 1) were synthesized as reported previously.[24]

Preparation of DN Gels. DN ionogels were synthesized in two steps. First, ionogels of low-molecular-weight gelators were prepared by heating solutions containing gelator **1** or **2** to 150 °C to dissolve the gelator and subsequent cooling to room temperature to allow the gelator molecules to self-assemble. The supramolecular gels were then immersed in [EtMeIm][TFSA] containing MMA (20 wt % to the solvent), EGDMA (2.0 mol % to MMA), and AIBN (2.0 mol % to MMA). After immersion for 24 h, the supernatant was removed, and polymerization was carried out at 60 °C for 24 h.

To estimate MMA content in the ionogel before polymerization, an ionogel of [EtMeIm][TFSA] (1 mL) and gelator **1** (1.0 wt %) was immersed in [EtMeIm][TFSA] (1 mL) containing MMA without EGDMA and incubated at room temperature for 24 h. The supernatant and ionogel were collected separately, and MMA was polymerized in both. After reaction at 60 °C for 24 h, PMMA was precipitated by adding an excess amount of methanol, dried under vacuum, and weighed to estimate the MMA content in the ionogel.

Preparation of PMMA Ionogels. Free radical polymerization was typically carried out by heating [EtMeIm][TFSA] containing MMA (20 wt % to the solvent), EGDMA (2.0 mol % to MMA), and AIBN (2.0 mol % to MMA) at 60 °C for 24 h.

Instrumentation. Molecular weights of PMMA were determined using a gel permeation chromatography system (Jasco LC2000 plus, Tokyo, Japan) equipped with two 80 mm × 300 mm columns (Showa Denko K.K., Tokyo, Japan) and a RI detector (Jasco RI2031 plus) at 40 °C. Chloroform was used as an elution solvent, and PMMA molecular weight standards were used for a calibration curve.

FE-SEM (JSM-7500F, JEOL, Tokyo, Japan) was operated at an accelerating voltage of 7 kV. Xerogels of the ionogels were prepared for FE-SEM following the procedure reported by Hanabusa et al.[20] Briefly, an ionogel was immersed in water for 3 days to exchange the ionic liquid for water, and the water was replaced several times. After removal of the ionic liquid, the gel was freeze-dried under vacuum to prepare the xerogel. The xerogel was mounted on an aluminum stub and coated with osmium by vapor deposition prior to analysis.

Confocal laser scanning microscope (CLSM) observations were carried out using a FV1000-D (Olympus Tokyo, Japan) at room temperature. The ionogel was stained with Rhodamine B (30 μM) for CLSM observation.

Elastic moduli of DN ionogels were measured using a dynamic viscoelastic analyzer (DVA-220S, ITK Co. Ltd., Osaka, Japan) with rectangular samples (5 × 10 × 1 mm) at room temperature. Samples were measured at a strain of 0.1% from a frequency of 1 to 110 Hz in shear mode.

Compressive stress−strain measurements were performed using a compressive tester (Autograph AGS-J, Shimadzu, Kyoto, Japan). A cylindrical gel sample with a diameter of 4 mm and a thickness of 2 mm was set on a lower plate and compressed by an upper plate, which was connected to a load cell, at a strain of 10% thickness/min.

Differential scanning calorimetry (DSC) measurements were carried out using a DSC 8500 (PerkinElmer, Waltham, MA, USA) with an aluminum pan at a heating rate of 10 °C/min.

Conductivities of the ionogels were measured at 25 °C using a Hioki Chemical Impedance Meter (3532-80; Ueda, Japan). A cylindrical gel sample with a diameter of 20 mm and a thickness of 2 mm was prepared to measure conductivity of an ionogel using a conductance cell (ECMC-P-20-POM, Eager Corp., Osaka, Japan).

ASSOCIATED CONTENT

Ⓢ Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsami.5b07981.

Full names of the ionic liquids and gelation results for other ionic liquids (PDF)

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Notes

The authors declare no competing financial interest.

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