Short communication

Binary ionic liquid electrolytes containing organo-magnesium complex for rechargeable magnesium batteries

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ABSTRACT

A binary ionic liquid (IL) system based on a common organic cation, *N,N*-diethyl-*N*-methyl-*N*-(2-methoxyethyl)anmmonium (DEME$^+$), with mixed anion of bis(trifluoromethanesulfonyl)imide (TFSI$^-$) and bis(fluorosulfonyl)imide (FSI$^-$) has been examined as a supporting electrolyte of rechargeable magnesium batteries. Basic properties of the mixed IL were measured as a function of the mixing ratio, *n*, in the composition of [DEME$^+$][TFSI$^-$]$_n$[FSI$^-$]$_{1-n}$. Electrochemical deposition/dissolution behavior of magnesium was investigated in the mixed IL containing an organo-magnesium complex, methyl magnesium bromide with tetrahydrofuran (MeMgBr/THF). Viscosity and ionic conductivity of the mixed IL showed monotonous changes with the mixing ratio, while their thermal properties proved discontinuous changes with the compositions. Coulombic (current) efficiency of 90% or higher was obtained for cathodic deposition of magnesium in the mixed IL containing MeMgBr/THF. The current density for cathodic deposition and anodic dissolution depended on the IL composition, and the maximum value was obtained at the composition of *n* = 0.5.

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1. Introduction

Magnesium (Mg) is an attractive candidate for the anode (negative electrode) active material of high energy-density batteries [1–5]. One of the key technologies for developing practical rechargeable batteries is establishing a reversible redox (deposition and dissolution) process of Mg. However, metal Mg is easily oxidized by water (moisture) and other protic-solvents to form passivation films [6,7]. Consequently, it is generally difficult to establish a reversible process of electrochemical deposition and dissolution of Mg, not only in aqueous but also in organic media. As a possible option to realize a reversible process of Mg deposition/dissolution, Liebenow [8] reported that a reversible redox process of Mg can be established in an ether solution of an organo-Mg complex, so-called Grignard Reagents. Since then, it has been generally accepted that no compact passivating film covers the Mg surface and then reversible deposition and dissolution of Mg occur with low overvoltage in such electrolyte systems as RMgX/ether (R: alkyl group).

On the other hand, ionic liquids (ILs), also known as room temperature molten salts, form a unique class of material. They are entirely composed of ions without solvents and in liquid states at ambient temperatures, which provide a great range of properties. Owing to their low vapor pressure, ILs are noted as novel reaction solvents [9] or green solvents [10,11]. Meanwhile, due to their high ionic conductivity, nonflammability and wide-electrochemical window, ILs have attracted much attention as novel ion conductive media for lithium-ion batteries [12,13], electrochemical capacitors [14,15], and dye-sensitized solar cells [16]. Applications as electrolytes in the lithium-ion and lithium-metal batteries are particularly promising as these properties enable the use of different cathode (positive electrode) materials of potentially higher energy density.

We have applied ILs as "ionic solvents" of the electrolytes containing organo-magnesium complexes, and demonstrated that the resulting Mg-complex/IL systems work as efficient electrolytes for reversible deposition/dissolution of Mg metal [17,18]. In those systems, the IL components suppress the vapor pressure and increase the ionic conductivity of the electrolyte system. As a result, higher efficiency for electrodeposition of Mg was achieved in a mixed system of organo-magnesium complex with IL than that in plain ether solutions of the organo-magnesium complex. In addition, as chemical properties of ILs are tunable with modifying their structure, we have many options when utilizing them in various areas of application. That is, according to the application area, we can design an IL with more desirable properties, including negligible vapor pressure, a wider electrochemical window and/or a wider temperature range of the liquid state. In this context, we have
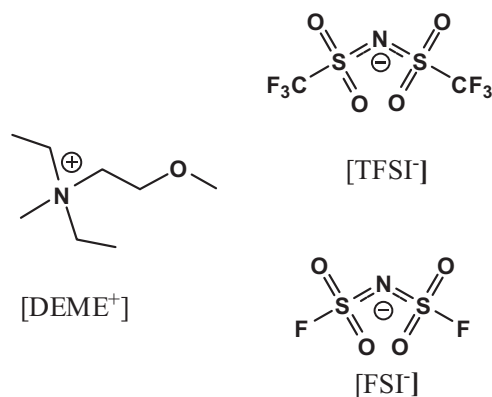

Fig. 1. Chemical structure of the ionic liquids.

successfully optimized the structure of imidazolium-based ILs as the ionic solvent for rechargeable Mg batteries [18]. We have demonstrated that high rechargeability of Mg depositon/dissolution can be achieved in the electrolytes using modified imidazolium cation, and that the chemical modification of ions in IL is effective to tune the electrochemical properties of the resulting electrolyte.

As the most ILs consist of organic cations with large-size anions, we have huge choices of the cation structures. By contrast, anions in ILs that have good chemical properties are limited. That is, the modification of anion structures is rather difficult, compared with the cationic species. Another approach to tune the anion properties is to use eutectic mixtures of two or more components that individually have attractive transport properties [19–23]. Numerous studies have been performed on binary or ternary IL systems, including molecular dynamics simulations and detailed studies on mixing and miscibility. In attempts to the electrochemical application, the composition ratio has not been studied enough. Most of the reported systems of mixed ILs change their properties linearly with the composition, while some systems showed local maximum/minimum values in certain mixing ratios. Thus, the mixing or blending of ILs having different chemical structures is effective to control or tune the ILs' properties.

In electrochemical applications, ILs based on N-alkyl-N-methyl-pyrrolidinium ($C_x$mpyr$^+$) or N,N-diethyl-N-methyl-N-(2-methoxyethyl)anmmonium (DEME$^+$) as the cation, with bis(trifluoromethanesulfonyl)imide (TFSI$^-$) or bis(fluorosulfonyl)imide (FSI$^-$) as the anion have so far been mainly used [24–27]. These classes of ILs are particularly promising for lithium battery applications as they can support lithium electrochemistry and help to form suitable solid electrolyte interphase (SEI), especially at carbon-based negative electrodes. That is, ILs consisting of TFSI$^-$ or FSI$^-$ have excellent properties such as liquidity over a wide temperature range, low viscosity and high ionic conductivity.

In the present study, we have chosen two ILs, DEMETFSI and DEMEFSI (Fig. 1) as the ionic solvent for Mg battery electrolytes. Some research groups have already examined mixed anion systems containing TFSI$^-$ and FSI$^-$ anions as lithium-ion battery electrolytes and demonstrated good rechargeability of graphite and silicon-based composite negative electrodes [26,28–30]. In this work, we have investigated basic electrochemical properties of the blended IL, [DEME$^+$][TFSI$^-$]$_n$[FSI$^-$]$_{1-n}$, as a function of the composition ratio, n. The electrochemical deposition and dissolution of Mg was examined in the blends dissolving an organo-magnesium complex, MeMgBr (Me = CH$_3$), to discuss the effect of anion mixing on the electrochemical behavior of Mg species in the Mg-complex/binary IL system.

2. Experimental

The component IL, DEMETFSI, used in this work was purchased from Kanto Chemical, which was dried at 80 °C under a vacuum before use. The other component DEMEFSI was synthesized and purified in our laboratory using the same methods as reported previously [31,32]. The chemical structure of the synthesized DEMEFSI was confirmed by ^{1}H NMR spectroscopy (JEOL, GSX-500). The water contents determined by Karl-Fischer titration (Kyoto Electronics, MKS-510) were approximately 200 ppm for all ILs after proper drying processes. An organo-magnesium complex, 1.0 mol dm^{-3} (M) of methyl magnesium bromide in THF (MeMgBr/THF, purchased from Kanto Chemical), was used as the Mg source. All reagents were stored in a glove box filled with dry Ar before use. Several compositions of binary ILs, [DEME$^+$][TFSI$^-$]$_n$[FSI$^-$]$_{1-n}$ (n = 0–1), were prepared by mixing each component in different molar ratios in the glove box. The Mg complex in THF was dissolved in the binary IL or each terminal component (DEMETFSI or DEMEFSI) with an equivalent amount of DEME$^+$ and Mg to form the electrolytic solution, MeMgBr/THF/IL ternary system.

Thermal properties of the electrolyte system were examined by DSC measurements (Shimadzu, DSC-50). The exo-/endothermic behavior was investigated for binary ILs and the ternary electrolytic solutions in the temperature range between −150 and 200 °C or −150 and 20 °C with a heating rate of 10 K min^{-1}.

The ionic conductivities of the binary ILs and the ternary electrolytic solutions were measured by an ac impedance technique using a frequency-response analyzer (FRA; Solartron, Type 1260) coupled with an electrochemical interface (Solartron, Type1287) over a frequency range from 10 to 10^6 Hz. The cathodic deposition and the anodic dissolution processes of Mg in the ternary electrolytic solutions, MeMgBr/THF/IL, were investigated by cyclic voltammetry (CV; Hokuto Denko, HZ-5000) using a conventional three-electrode beaker cell. A nickel (Ni) disc was used as the working electrode (substrate). A large surface area Pt sheet and Mg wire were employed as a counter and a quasi-reference electrode (QRE), respectively. The potential scan rate for CV was 10 mV s^{-1}. Charge (deposition) and discharge (dissolution) characteristics were examined under a constant-current condition using the same three-electrode cell as used in CV, except for an Au sheet as the working electrode (substrate). The cell was cycled with a current density of 5 mA cm^{-2} and the cut off condition of 120 s for charging and 1.5 V (vs. Mg QRE) for discharging. These electrochemical experiments including electrolyte preparation were carried out under a dry Ar atmosphere at room temperature (22 ± 3 °C). The deposit on the Ni disk substrate after constant-potential polarization was analyzed by an X-ray diffractometer (XRD; Rigaku, ATX-G) and a scanning electron microscope (SEM; KEYENCE, VE-8800).

3. Results and discussion

The ionic conductivities of binary ILs and their terminal component ILs, DEMETFSI and DEMEFSI, were determined by the ac impedance method. Fig. 2 shows temperature dependence of the ionic conductivity of the binary ILs, where the composition is expressed as [DEME$^+$][TFSI$^-$]$_n$[FSI$^-$]$_{1-n}$ (n = 0–1). Arrhenius-type plots of the conductivity (σ_i) exhibit curved profiles without discontinuous transitions. At room temperature (∼25 °C), the conductivity showed a maximum value, ca. 10 mS cm^{-1}, for DEMEFSI, i.e. the end composition of n = 0. At higher temperature, however, the composition of n = 0.3 gave the highest conductivity among the system examined. These suggest that the activation energy for ionic conduction depends on the composition. The ionic conductivity at and below room temperature decreased with increasing the TFSI$^-$ content, n. As shown in Fig. 3, it varied almost linearly between the end compositions, DEMEFSI and DEMETFSI, at room temperature.
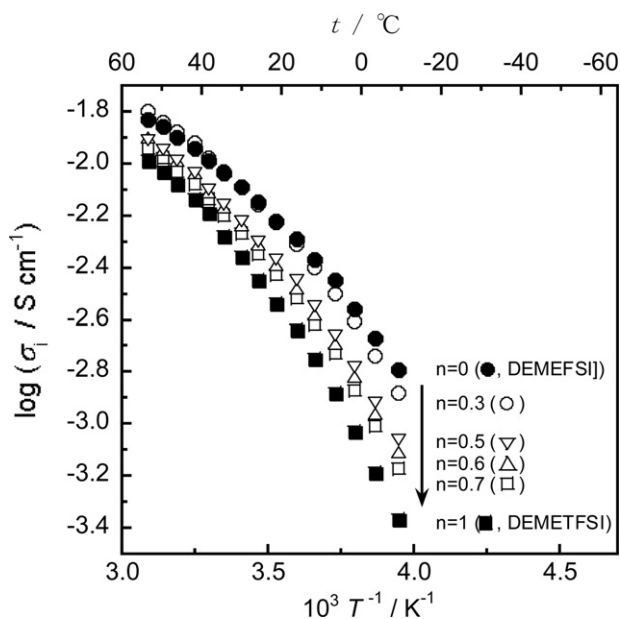
Fig. 2. Temperature dependence of the ionic conductivity (σ_i) for the binary ionic liquid system, $[DEME^+][TFSI^-]_n[FSI^-]_{1-n}$.

Fig. 4. Ionic conductivity of the electrolyte solution (σ_i) at 25 °C as a function of the IL composition (n) for $MeMgBr/THF/[DEME^+][TFSI^-]_n[FSI^-]_{1-n}$.

The ionic conductivity (σ_i) in condensed materials is generally described as Eq. (1),

$$\sigma_i = \sum (m \times e \times \mu) \tag{1}$$

where m, e, and μ are the number of ionic species, the elementary charge, and the mobility of ions, respectively. In the present system, the same amounts of the cation ($DEME^+$) and the anions ($TFSI^- + FSI^-$) exist in every electrolyte composition. If the degree of ion-dissociation does not change intrinsically with the mixing ratio of the anion, the ionic conductivity of the system will depend on the mobility of each ion. The viscosity of the mixed IL decreased with the FSI^- content, n, in the system. Thus, the main cause of the changes in the ionic conductivity with the composition would be the changes in the viscosity of the mixed IL.

Thermal properties of the binary IL, $[DEME^+][TFSI^-]_n[FSI^-]_{1-n}$, were examined by DSC. Each composition showed clear melting point (T_m) and glass transition temperature (T_g). In Fig. 3, T_g of each
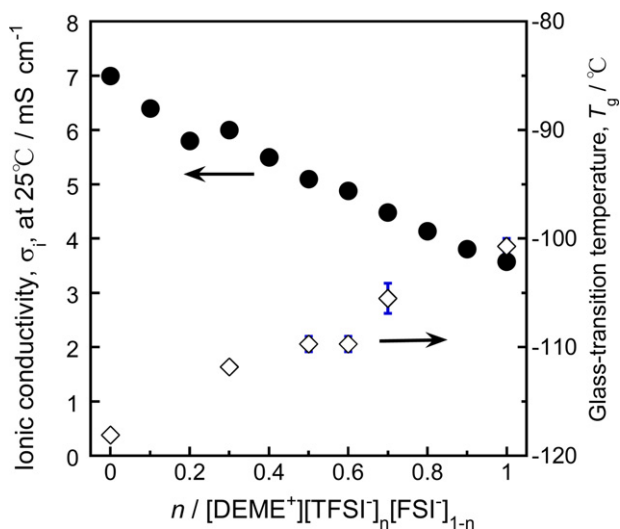

Fig. 3. Variations in the ionic conductivity (σ_i) at 25 °C and the glass-transition temperature (T_g) with the composition ratio (n) for $[DEME^+][TFSI^-]_n[FSI^-]_{1-n}$.

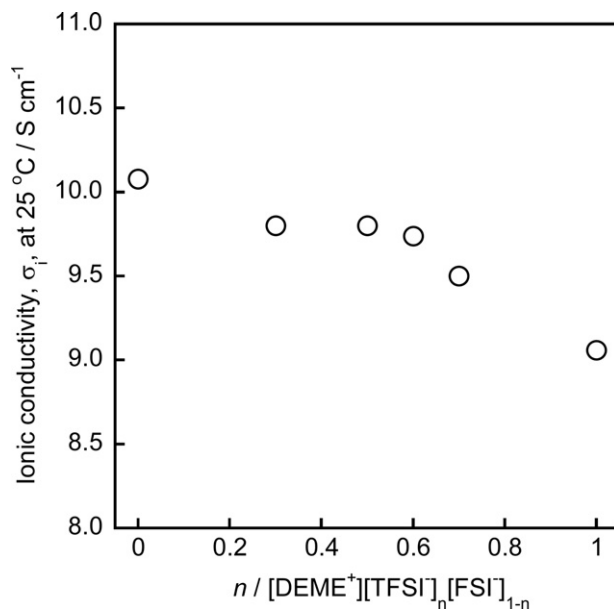system is also shown as a function of the mixing composition, n. For the present binary system, T_g varies almost linearly with the composition ratio, and DEMEFSI showed the lowest T_g. It is clear that the variation in T_g obtained from the DSC measurement corresponds to that in the ionic conductivity. That is, the ionic conductivity in the present binary IL is determined by its thermal property, through the viscosity. These results strongly suggest that differences in structure and properties of the ions, $TFSI^-$ and FSI^-, lead to different ionic interactions between $DEME^+$ cation and the anions.

The organo-magnesium complex, MeMgBr in THF (MeMgBr/THF), as a magnesium source was dissolved in $DEME^+$-based ILs with mixed anions of $TFSI^-$ and FSI^-. The ionic conductivity of the obtained electrolyte solution, $MeMgBr/THF/[DEME^+][TFSI^-]_n[FSI^-]_{1-n}$, was measured by the ac impedance method. Fig. 4 shows the obtained conductivity as a function of the composition ratio, n, in $[DEME^+][TFSI^-]_n[FSI^-]_{1-n}$ at 25 °C. As expected, the conductivity of $MeMgBr/THF/[DEME^+][TFSI^-]_n[FSI^-]_{1-n}$ was higher than that of IL without the magnesium complex, shown in Fig. 3. This is because the viscosity of the electrolyte solution decreasing much by the addition of low viscosity THF, as the solvent for MeMgBr. Thus, the ionic conductivity itself is higher and its composition dependence becomes smaller than those observed in the binary IL without MeMgBr/THF.

The thermal properties of the Mg-complex/binary IL system were also examined by DSC. Fig. 5 shows the DSC profiles of the electrolyte solutions based on binary IL. Below $n = 0.5$, a phase-transition (T_g) was clearly observed in each DSC curve and its value shifted to lower temperature with the increase in the composition, n. However, the system whose composition ratio was higher than 0.5, T_g became unclear and exothermic peak derived from another phase-transition, probably melting (T_m), appeared at around −60 °C. These phenomena were not observed in the binary ILs without the Mg-complex. Thus, it is suggested that some heterogeneity of ions increases with the increase in the $TFSI^-$ content in the electrolyte solutions containing the Mg-complex for the anion content of $TFSI^-$ (n) being higher than 0.5. The mobility of molecular components, especially MeMgBr and THF, would increase with the heterogeneity of ions corresponding to the decrease in T_g. On the other hand, in the electrolyte solutions with lower content of
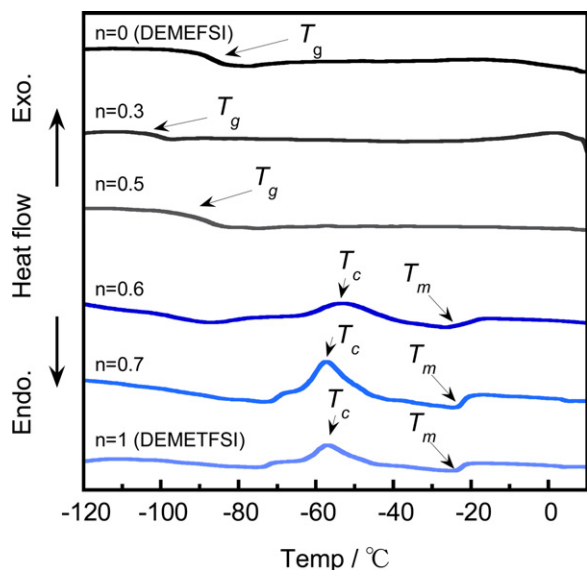
Fig. 5. DSC curves for the ternary system, MeMgBr/THF/[DEME$^+$][TFSI$^-$]$_n$[FSI$^-$]$_{1-n}$, obtained at the second heating cycle.

Fig. 7. Quantity of electricity passed for anodic Mg dissolution (Q_{dis}) as a function of the composition ratio (n) in MeMgBr/THF/[DEME$^+$][TFSI$^-$]$_n$[FSI$^-$]$_{1-n}$.

TFSI$^-$ (e.g. $n < 0.5$) in MeMgBr/THF/[DEME$^+$][TFSI$^-$]$_n$[FSI$^-$]$_{1-n}$, the DSC results imply that the degree of crystallization will increase with the increase in the TFSI$^-$ content due to its larger ion size than FSI$^-$.

Cyclic voltammetry (CV) was conducted in the electrolyte solutions of MeMgBr dissolved in [DEME$^+$][TFSI$^-$]$_n$[FSI$^-$]$_{1-n}$. Fig. 6 shows typical CV curves which were obtained for an Ni disc electrode in the electrolyte solution based on DEMEFSI as IL ($n = 0$). Stable voltammetric response was observed in this solution at the second cycle and after it. The cathodic current increase from -0.3 to -1.2 V (vs. Mg QRE) corresponds to Mg deposition at the Ni substrate, and the anodic peak at around 1.0 V is derived from the electrochemical dissolution of deposited Mg. Similar current responses to that shown in Fig. 6 were obtained for the electrolyte systems with all compositions of the binary IL. In Fig. 7, the quantity of electricity passed for the anodic Mg dissolution (Q_{dis}), observed in its third cycle, is plotted as a function of the IL composition. As clearly seen, the electrolytes composed of binary ILs showed higher electricity, ie. higher amount of Mg deposition, than the
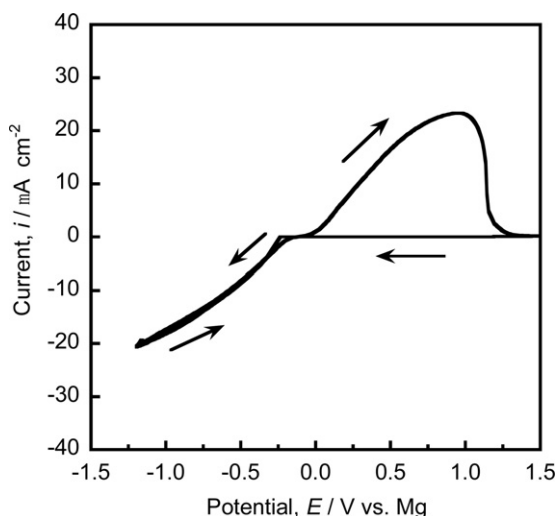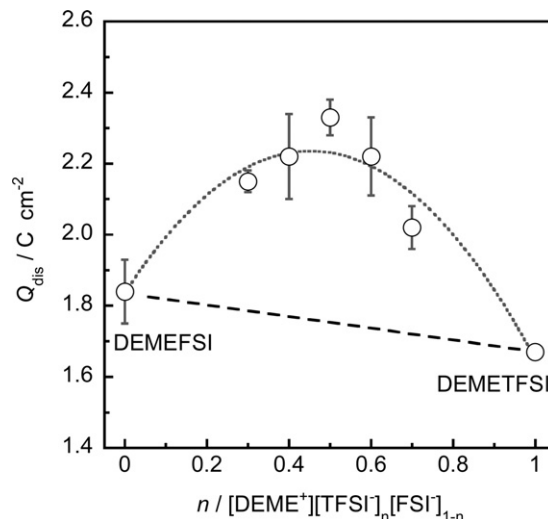
electrolyte consisting of single IL, DEMEFSI or DEMETFSI. The highest electricity was obtained in the electrolyte solution composed of [DEME$^+$][TFSI$^-$]$_{0.5}$[FSI$^-$]$_{0.5}$. The voltammetric response in the IL composition of $n = 0.5$, shown in Fig. 8, also leads to the lowest overvoltage for the Mg deposition/dissolution, which is typically revealed by higher peak current and lower peak potential (ca. 0.5 V) for the anodic reaction. This is probably because the electrolyte composition with $n = 0.5$ would have a preferable ionic structure to proceed the electrochemical redox reaction of Mg on the Ni substrate.

Constant-potential electrodeposition was conducted on the Ni disc electrode in the electrolyte consisting of MeMgBr/THF/[DEME$^+$][TFSI$^-$]$_{0.5}$[FSI$^-$]$_{0.5}$ at -0.5 V vs. Mg QRE. A grayish uniform deposit was formed on the Ni substrate after 180 min of the electrolysis. The deposit was rinsed with THF in an Ar atmosphere and then dried at room temperature in-vacuo. The XRD pattern of the resulting deposit is shown in Fig. 9. The diffraction peaks observed at $2\theta = 32.2°$, $34.4°$, $36.6°$, $57.4°$, and $63.1°$ are attributed to the hexagonal Mg-metal phase according to the database of JCPDS. Surface image of the deposit by scanning electron micrograph (SEM) is shown in the inset of Fig. 9. The deposit
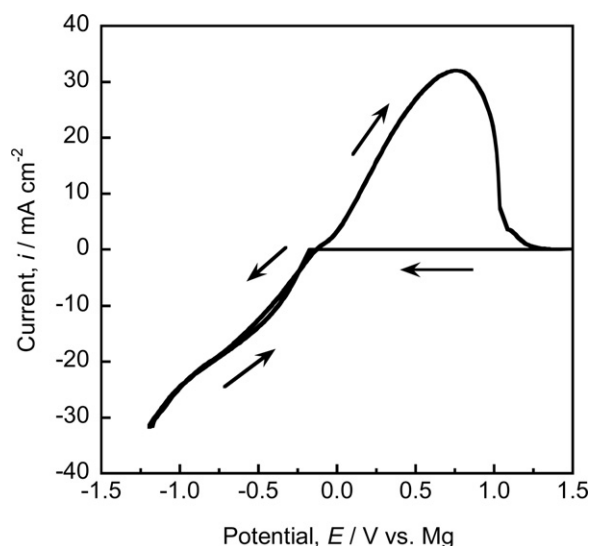

Fig. 6. Cyclic voltammogram for an Ni substrate in the electrolyte solution of MeMgBr/THF/DEMEFSI.

Fig. 8. Cyclic voltammogram for an Ni substrate in the electrolyte solution of MeMgBr/THF/[DEME$^+$][TFSI$^-$]$_{0.5}$[FSI$^-$]$_{0.5}$.
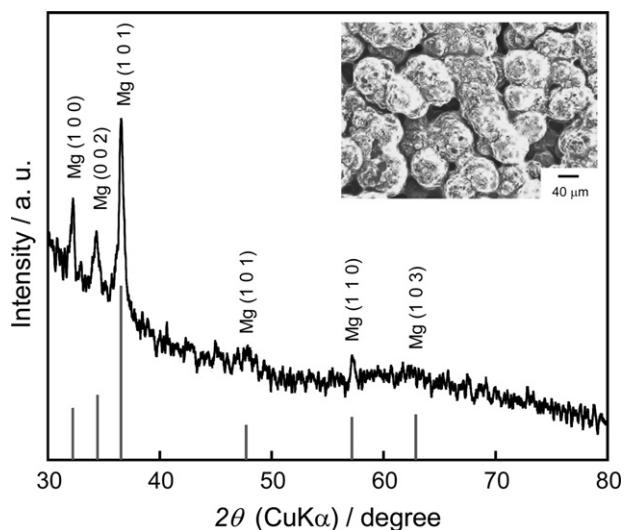
Fig. 9. XRD pattern and SEM image of the electrodeposit on an Ni substrate from the electrolyte solution of MeMgBr/THF/[DEME$^+$][TFSI$^-$]$_{0.5}$[FSI$^-$]$_{0.5}$.

consisted of solid particles with approximately 50 μm diameter. These results proved that the present Mg-complex/binary IL system works as an efficient electrolyte for cathodic deposition of Mg metal.

Constant-current deposition/dissolution of Mg was also cycled in the solution containing MeMgBr/[DEME$^+$][TFSI$^-$]$_{0.5}$[FSI$^-$]$_{0.5}$. Coulombic efficiency for charge and discharge (deposition/dissolution) cycle was determined from each potentiogram. Fig. 10 shows the efficiency variation with the cycle number under the current density of ±5 mA cm^{-2}. About 90% or higher efficiency was achieved over long cycles (>100 cycles) in the present organo-Mg complex in binary IL, MeMgBr/THF/[DEME$^+$][TFSI$^-$]$_{0.5}$[FSI$^-$]$_{0.5}$. Thus, we believe that further optimization of the ionic structure of Mg-complex/IL system will realize a promising electrolyte system for rechargeable Mg battery.
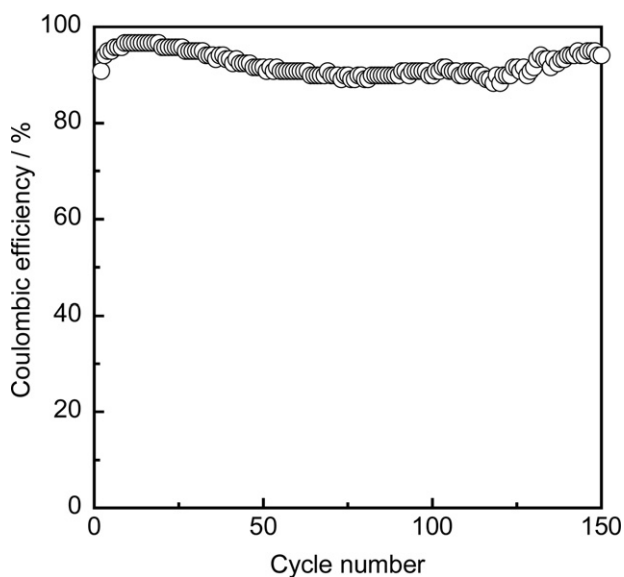

Fig. 10. Variation in the coulombic efficiency with the cycle number for Mg deposition/dissolution process in the electrolyte solution of MeMgBr/THF/[DEME$^+$][TFSI$^-$]$_{0.5}$[FSI$^-$]$_{0.5}$.

4. Conclusions

A binary IL system consisting of a common cation, DEME$^+$, combined with different structures of anions, TFSI$^-$ and FSI$^-$, has been examined as an ionic solvent for rechargeable Mg batteries. Effects of the anion blending on the electrochemical behavior of the ionic liquid itself and the solution dissolving organo-Mg complex have been investigated, and the results are summarized as follows:

1. Ionic conductivity of the binary IL, [DEME$^+$][TFSI$^-$]$_n$[FSI$^-$]$_{1-n}$, showed monotonous change with the mixing ratio of TFSI$^-$ and FSI$^-$, which corresponds to the change in the viscosity of the system.
2. Thermal behavior of the binary IL indicated gradual change in glass-transition temperature. The DSC responses of the electrolyte solutions containing the Mg complex showed small changes in the ionic structure at the composition around $n = 0.5$, where the phase-transition mode was changed from glass-transition to melting.
3. Voltammetric responses showed good reversibility for Mg deposition and dissolution in the electrolyte solution of MeMgBr/THF dissolved in the binary IL. The highest current density for the dissolution was observed in the composition of $n = 0.5$.
4. About 90% or higher coulombic efficiency was achieved for charge/discharge (deposition/dissolution) processes under constant-current conditions over 100 cycles or more, which suggests that the present Mg-complex/binary IL system can work as an efficient electrolyte for rechargeable Mg batteries.

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