Ionics
DOI 10.1007/s11581-016-1870-3

ORIGINAL PAPER

Polyethylene oxide and ionic liquid-based solid polymer electrolyte for rechargeable magnesium batteries

H. N. M. Sarangika[1,2,3] · **M. A. K. L. Dissanayake**[2,3] · **G. K. R. Senadeera**[2,4] · **R. R. D. V. Rathnayake**[5] · **H. M. J. C. Pitawala**[5]

Received: 21 August 2016 / Revised: 6 October 2016 / Accepted: 13 October 2016

Abstract Solid polymer electrolytes (SPEs) based on polyethylene oxide (PEO) complexed with magnesium triflate $Mg(Tf)_2$ or $Mg(CF_3SO_3)_2$) and incorporating the ionic liquid (IL) (1-butyl-1-methylpyrrolidinium bis(trifluoromethanesulfonyl)imide ($PYR_{14}TFSI$)) were prepared by solution cast technique. The electrolyte was optimized and characterized using electrical conductivity, cationic transport number measurements, and cyclic voltammetry. The highest conductivity of the PEO/$Mg(Tf)_2$, 15:1 (molar ratio), electrolyte at room temperature was 1.19×10^{-4} S cm^{-1} and this was increased to 3.66×10^{-4} S cm^{-1} with the addition of 10 wt.% ionic liquid. A significant increase in the Mg^{2+} ion transport number was observed with increasing content of the ionic liquid in the PEO-$Mg(Tf)_2$ electrolyte. The maximum Mg^{2+} ion transport number obtained was 0.40 at the optimized electrolyte composition. A battery of the configuration Mg/ and $[(PEO)_{15}$:$Mg(Tf)_2$+10%IL]/TiO_2-C was assembled and characterized. Preliminary studies showed that the discharge capacity of the battery was 45 mA h g^{-1}.

Keywords PEO · $Mg(Tf)_2$ · Ionic liquid · Solid polymer electrolyte · Rechargeable Mg ion batteries

Introduction

In the energy storage scenario, rechargeable batteries appear to offer the best solutions available at present. In the field of modern advanced power sources, lithium-ion (Li-ion) batteries are the systems of choice, offering high energy density, flexible and lightweight design, and longer lifespan than comparable battery technologies. The major challenges for developing Li-ion batteries are performance, safety, and cost. Problems such as dendrite deposition of lithium and growth of passive layer on lithium surface lead to poor rechargeability of the battery. Additionally, lithium is highly reactive towards atmospheric oxygen and moisture, contributing to safety and environmental concerns [1, 2].

Magnesium is positioned next to the lithium in the periodic table and its electrochemical properties are comparable with Li. Therefore, Mg is an attractive candidate for the anode material of high energy density Mg batteries. However, Mg deposition is impossible in some electrolytes containing simple Mg ionic salts like $MgCl_2$ and $Mg(ClO_4)_2$ in commonly used aprotic solvents such as alkyl carbonates and esters because of formation of passive layers [3]. Surface film which covers Mg electrode can block the divalent Mg^{2+} ions. Polymer or gel polymer electrolytes might be useful to overcome this problem.

Magnesium ion-conducting gel polymer electrolytes for magnesium-ion rechargeable batteries have received a particular interest because of advance performances in charging and discharging, power density, and high volumetric capacity. These electrolytes are also much safer,

---

✉ H. N. M. Sarangika
sarangikah@gmail.com

[1] Department of Physical Sciences and Technology, Sabaragamuwa University of Sri Lanka, Belihuloya, Sri Lanka

[2] National Institute of Fundamental Studies, Kandy, Sri Lanka

[3] Postgraduate Institute of Science, University of Peradeniya, Peradeniya, Sri Lanka

[4] Department of Physics, The Open University of Sri Lanka, Nawala, Nugegoda, Sri Lanka

[5] Faculty of Science and Technology, Uva Wellassa University, Badulla, Sri Lanka

less reactive, and easier to handle in ambient temperature compared to lithium systems. However, it would be desirable to use non-volatile electrolytes with low flammability for development of practical Mg batteries. Ionic liquid-based electrolytes possess many advantages such as high ionic conductivity, wide electrochemical window, greater thermal and chemical stability, non–flammability, and non-volatility. However, there are so far very few results reported on the ionic liquid (IL)-based electrolytes for Mg^{2+} ion conduction. Performance of reversible deposition and dissolution of Mg in imidazolium-based ionic liquid with Grignard reagents have been reported [4]. Nabuko et al. [5] reported Mg electrodeposition from the electrolyte containing ammonium-based ionic liquid with Grignard reagents. Few magnesium ions conducting gel polymer electrolytes are reported in which the liquid component is much higher (~80 wt.%) with respect to host polymers like PVdF-HFP and PEO-PMA [6, 7]. All these studies have mainly focused on the performance of IL-based gel polymer electrolytes in batteries and characterization of transport properties. However, in polymer electrolyte system, incorporation of IL of 5–40 wt.% is also beneficial to enhance the electrochemical properties. Such ionic liquid system is basically semisolid in which the solid polymer network having ionic liquid region entrapped. By incorporating a suitable IL into such a semisolid electrolyte system, the battery performance can be improved in terms of capacity and cyclability [8]. These semisolid or solid electrolyte membranes are free-standing, flexible, and non-volatile. Several problems associated with the conventional liquid electrolytes, such as flammability and volatility that limits the safety and reliability of the battery, can be overcome by replacing the traditional electrolytes with the electrolyte based on ILs [9, 10].

Among different polymers used, polyethylene oxide (PEO) is more promising than other polymers due to its high solvation ability. This is mainly due to its ability to dissolve salts [$LiClO_4$, $LiBF_4$, $LiCF_3SO_3$, $NaClO_4$, $Mg(ClO_4)_2$, etc.] well and proper chemical structure to support cation transport. In this work, we report novel magnesium ion-conducting solid polymer electrolyte system based on room temperature ionic liquid that consists of 1-butyl-1-methylpyrrolidinium bis(trifluoromethanesulfonyl)imide ($PYR_{14}TFSI$) magnesium triflate or $Mg(Tf)_2$ and polyethylene oxide. The electrolyte consists of least amount of IL (10 wt.%) which has not been reported and shows high conductivity and magnesium transport number. A wide variety of experiments, namely, complex impedance analysis, conductivity measurements, transference number, and cyclic voltammetry, have been carried out to characterize the solid polymer electrolyte and to establish ion transport behavior of the electrolyte.

Materials and methods

Materials

PEO (M_w400000) and magnesium trifluoromethanesulfonate ($Mg(CF_3SO_3)_2$) (abbreviated as magnesium triflate, $Mg(Tf)_2$ and 1-butyl-1-methylpyrrolidinium bis(trifluoromethanesulfonyl)imide ($PYR_{14}TFSI$) >98 % purchased from Sigma Aldrich, acetonitrile with purity >98 % purchased from Fluka, and fluorine-doped SnO_2-layered (FTO) glass (sheet resistance 12 Ω/sq) and TiO_2 (Degussa P-25) purchased from Solaronix SA were used as staring materials. PEO was vacuum dried at ~50 °C, whereas the salt $Mg(Tf)_2$ was dried at ~100 °C in vacuum for 24 h before their use to prepare polymer electrolytes.

Preparation of electrolyte

PEO was vacuum dried at 50 °C and $Mg(Tf)_2$ was vacuum dried at 100 °C for 24 h. The polymer electrolytes were prepared by “solution-cast” technique. In this process, the PEO was first dissolved in acetonitrile by stirring it magnetically at room temperature (~25 °C). The electrolyte films having different molar ratios of PEO/$Mg(Tf)_2$ [10:1, 15:1, 20:1, 25:1, and 30:1] were made by common solvent casting method and vacuum dried at 50 °C for 5 to 6 h. Ionic conductivity was calculated for each sample, and sample with the highest ionic conductivity was optimized for ionic liquid treatment. The ionic liquid-based samples were prepared after the addition of x wt.% IL (x = 5, 10, 15, 20) into the electrolyte which shows high ionic conductivity. The solution was poured onto the aluminum foil covered by the glass ring and vacuum dried at 50 °C for 5–6 h in order to obtain a free-standing solid polymer electrolyte (SPE) film.

Preparation of electrode

A powder mixture (0.2 g) of 89 wt.% TiO_2, 0.025 g of 11 wt.% carbon powders, one drop of Triton-X (as a binder), and few drops of acetic acid were thoroughly ground with ethanol. This mixture was used to prepare the cathode pellets with 5 mm diameter and thickness 12 μm on FTO glass substrate and then sintered at 450 °C for 45 min. Since we need to minimize the amount of binder used in the cathode, we followed this unconventional method to prepare the TiO_2 cathode on different current collectors (FTO). Sintering up to 450 °C completely removes the binder (Triton-X) used in preparing the cathode and this will help to decrease the amount of binder used in preparing cathode as well as internal resistance of the battery and hence increase the battery capacity. It is impossible to remove binder by sintering with the use of other commonly used current collectors such as Cu and Al. The weight of the prepared TiO_2 electrode was in the 1–2-mg range out of which the weight of the active TiO_2 material was 89 %. Pure Mg disc was used as the anode. Thin, circular discs of magnesium metal were sectioned out of a Mg sheet, polished and cleaned using acetone. The weight of the magnesium discs used as the anode was about 0.15 g.

Instrumentation

The ionic conductivity of electrolyte samples was determined using impedance measurements by a computer-controlled Metrohm Autolab (PGSTAT 128N) impedance analyzer in the frequency range from 0.1 Hz to 10 MHz. For impedance measurements, a disc-shaped solid polymer electrolyte sample was sandwiched between two polished stainless steel (SS) electrodes with the configuration SS/SPE/SS. The sample assembly was placed in a furnace with a temperature controller, and impedance measurements were taken from room temperature up to 65 °C at 5 °C intervals. Conductivity values at each temperature were extracted from the impedance measurements.

The electronic contribution to the total ionic conductivity was estimated by the DC polarization measurements. In this method, DC polarization current was monitored as a function of time for the electrolyte sample sandwiched between two stainless steel blocking electrodes with the configuration SS/SPE/SS and by applying a 1 V DC across the sample [11].

The total ionic transference number (t_{ion}) of the polymer electrolyte was estimated by Wagner's method using the equation

$$t_{\text{ion}} = \frac{I_i - I_f}{I_i} \qquad (1)$$

where I_i is the initial current and I_f is the final steady state current.

In order to determine the Mg^{++} ion contribution to the total ionic conductivity, a combination of AC and DC measurements were taken. The method suggested in the literature [11–14] was used to calculate the magnesium ion (Mg^{2+}) transference number (t_{Mg2+});

$$t_{\text{Mg++}} = \frac{I_s(\Delta V - r_0 I_0)}{I_0(\Delta V - r_s I_s)} \qquad (2)$$

where I_0 and I_s are the initial and final steady state currents, and r_0 and r_s are the cell resistance before and after applying the polarization voltage, respectively [12]. For finding the $r_0$value, the electrolyte was sandwiched between two non-blocking Mg electrodes with the cell configuration Mg/SPE/Mg, and the complex impedance response of the symmetrical cell assembly was measured. As followed by the literature [11–14], a small voltage pulse (ΔV = 0.3 V) was applied to the cell until the polarization current reached a steady state after which the complex impedance was again measured.

Cyclic voltammograms of the symmetrical cells SS/SPE/SS and Mg/SPE/Mg were recorded using a computer-controlled Metrohm Autolab potentiostat galvanostat (PGSTAT 128N) at 5 and 1 mV s^{-1} scan rates. The experiments were carried out at 30 °C.

The Mg/[$(PEO)_{15}$:$Mg(Tf)_2$+10%IL]/TiO_2-C cell was assembled using the anode, electrolyte, and cathode. Charge/discharge cycling characteristics of the cells were recorded at room temperature using the Metrohm Autolab potentiosat/galvanostat (PGSTAT 128N) system.

Results and discussion

Characterization of the gel polymer electrolyte

Electrical conductivity

First, the ionic conductivity of the electrolyte samples having different molar ratio of PEO/$Mg(Tf)_2$ was measured. Figure 1a shows the temperature dependence of ionic conductivity of studied samples with different PEO/$Mg(Tf)_2$ molar ratios. The linear shape of the graph suggests that the temperature dependence of the conductivity obeys the classical Arrhenius relation expressed by the equation $\sigma = \sigma_o \exp(-E_a/RT)$. Among the studied samples, PEO/$Mg(Tf)_2$, 15:1, sample shows the highest ionic conductivity with the value of 1.19×10^{-4} S cm^{-1} at room temperature. Figure 1b shows the influence of ionic conductivity of parent electrolyte [$(PEO)_{15}$:$Mg(Tf)_2$] with the addition of different amounts of ionic liquid PYR_{14}TFSI. Our results indicate a clear conductivity enhancement with the addition of 10 % PYR_{14}TFSI into PYR_{14}TFSI-free electrolyte giving the highest value of 3.66×10^{-4} S cm^{-1} at room temperature. However, on further addition of PYR_{14}TFSI, although the ionic conductivity increases, the electrolyte becomes mechanically liquid-like and the gain in conductivity enhancement is also not that much significant at room temperature. The possible reasons of enhancement of ionic conductivity are improved flexibility in the polymer backbone and the introduction of additional PYR_{14}^+ mobile charge species into the electrolyte due to the incorporation of high dielectric constant ionic liquid [11].

The ionic transference number of the electrolyte was estimated from DC polarization measurements. Results of the DC polarization current measured as a function of time on application of DC potential of 1.0 V across the cell with SS blocking electrodes in configuration SS/SPE/SS show that the polarization current dropped by more than 97 % from the initial value during the first 20 min and then gradually became steady after about 3 h (Fig. 2). Table 1 shows the summarized results of DC polarization test and it shows clearly that the electronic transference number has decreased considerably due to addition of PYR_{14}TFSI. This further indicates

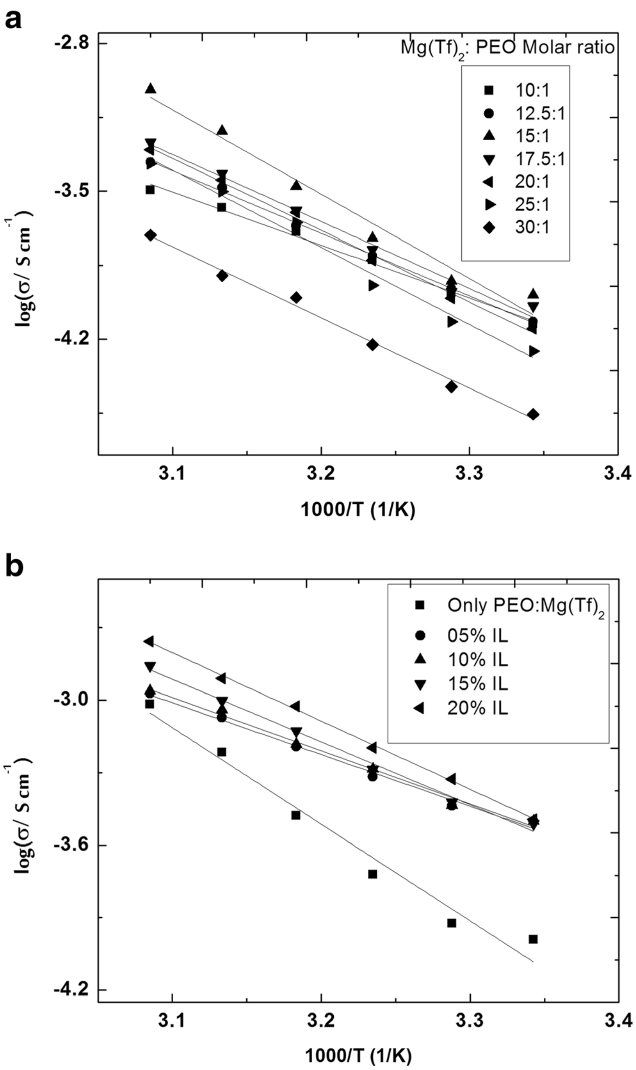

Fig. 1 The variation of ionic conductivity vs inverse temperature for studied polymer electrolytes of **a** IL-free electrolytes and **b** IL-added electrolytes

that the ionic nature of the electrolyte has been improved due to the IL.

The total ionic transference number, t_{ion}, estimated using the equation $t_{\mathrm{ion}} = (I_i - I_f) / I_i$ is 0.976 for [(PEO)$_{15}$:Mg(Tf)$_2$+10%IL]. This shows that the material is predominantly an ionic conductor, with Mg^{2+}, $PYR_{14}{}^{+}$, and $CF_3SO_3{}^{\mathbf{2-}}$ as the dominant mobile ionic species. The total ionic conductivity of the electrolyte is made up of individual ionic conductivity contributions from these three ionic species. However, Mg^{2+} ion contribution for the total ionic conductivity is important for the Mg battery, because Mg^{2+} ions intercalate/de-intercalate into/from TiO_2 cathode during the charging/discharging of the battery.

In order to determine the Mg^{2+} ion transference number, AC impedance of the Mg/SPE/Mg symmetrical cell with the electrolyte samples [(PEO)$_{15}$:Mg(Tf)$_2$] and [(PEO)$_{15}$:Mg(Tf)$_2$+10%IL] was measured. Subsequently, a DC polarization voltage of $\Delta V = 0.30$ V was applied across the symmetrical cell Mg/SPE/Mg, and the initial current I_0 and resulting steady state current I_s were recorded. Figure 3 shows the variation of DC polarization current as a function of time.

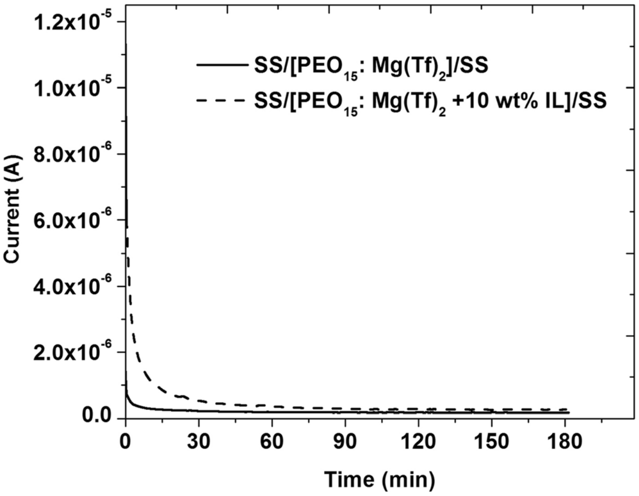

Fig. 2 The DC polarization curves of parental electrolyte [PEO$_{15}$:Mg(Tf)$_2$], electrolyte incorporating 10 % of IL [PEO$_{15}$:Mg(Tf)$_2$+10 wt.% IL] with stainless steel (SS) blocking electrodes

According to the DC polarization measurements, the Mg^{2+} ion transference number for electrolyte [(PEO)$_{15}$:Mg(Tf)$_2$] is 0.21 and for [(PEO)$_{15}$:Mg(Tf)$_2$+10%IL] is 0.40. This shows that the Mg^{2+} ion contribution to the total ionic conductivity is significant for the electrolyte incorporated with PYR_{14}TFSI.

In order to confirm the Mg^{2+} ion conduction in [(PEO)$_{15}$:Mg(Tf)$_2$+10%IL] electrolyte system, impedance spectroscopy and CV measurements were performed. Figure 4 shows the comparative impedance plots for the symmetrical cells consisting of [(PEO)$_{15}$:Mg(Tf)$_2$+10%IL] solid polymer electrolyte with blocking (SS) and non-blocking (Mg) electrodes. Steep rising impedance pattern of SS/[(PEO)$_{15}$:Mg(Tf)$_2$+10%IL]/SS symmetrical cell indicates the ion blocking nature of SS electrodes to the Mg^{2+} ions at the electrode electrolyte interfaces. Semicircular dispersion curve obtained for the Mg/[(PEO)$_{15}$:Mg(Tf)$_2$+10%IL]/Mg shows the reversible nature of the Mg electrodes in the polymer electrolyte.

Table 1 Electronic transference number of [PEO$_{15}$: Mg(Tf)$_2$] and [PEO$_{15}$: Mg(Tf)$_2$ + 10 wt.% IL] samples

Sample	Initial current (I_0)	Steady current (I_s)	Electronic transference number	Cationic transference number
[PEO$_{15}$: Mg(Tf)$_2$]	1.4×10^{-6} A	1.8×10^{-7} A	0.129	0.19
[PEO$_{15}$: Mg(Tf)$_2$ + 10 wt% IL]	1.14×10^{-5} A	2.7×10^{-7} A	0.024	0.40

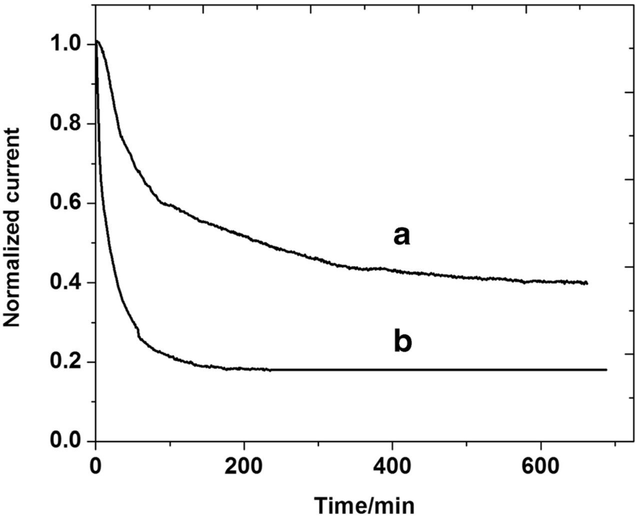

Fig. 3 Variation of the DC polarization current (normalized) as a function of time for the (*a*) Mg/[$(PEO)_{15}$:$Mg(Tf)_2$ + 10 %PYR_{14}TFSI]/Mg cell and (*b*) Mg/[$(PEO)_{15}$:$Mg(Tf)_2$]/Mg cell

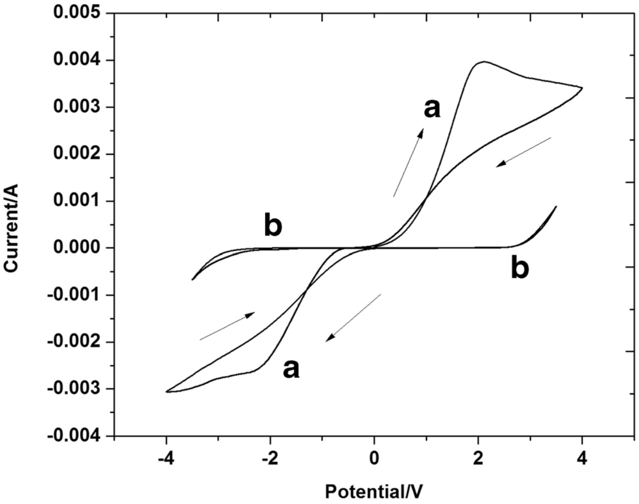

Fig. 5 Cyclic voltammograms of (*a*) Mg/[$(PEO)_{15}$:$Mg(Tf)_2$+ 10%PYR_{14}TFSI]/Mg symmetrical cell and (*b*) SS/[$(PEO)_{15}$:$Mg(Tf)_2$+ 10%PYR_{14}TFSI]/SS symmetrical cell taken at a scan rate of 5 mV s^{-1} and at 30 °C

Similar results have been reported by Kumar Y., et al. [11] for the PEO-based electrolyte containing EMITf ionic liquid.

Electrochemical stability and reversibility of magnesium in [$(PEO)_{15}$:$Mg(Tf)_2$+10%PYR_{14}TFSI]-based electrolyte is followed by the cyclic voltammetry with symmetrical non-blocking magnesium electrodes. The Mg/SPE/Mg symmetrical cells were subjected to cyclic voltammetry with the scan rate of 5 mV s^{-1} and voltagramm is shown in Fig. 5. The cyclic voltammetry of SS/SPE/SS blocking symmetrical cell has also been taken and shown in the Fig. 5 for the comparison. The cathodic and anodic current peaks are clearly observed for the Mg/SPE/Mg symmetrical cell. Higher magnitude of cathodic and anodic peak currents of the cell Mg/GPE/Mg suggest that the cathodic deposition and anodic oxidation occur at the Mg/SPE interface according to the reaction given below by Eq. ((3) [1]:

$$Mg^{+2} + 2e^{-} \leftrightarrow Mg \tag{3}$$

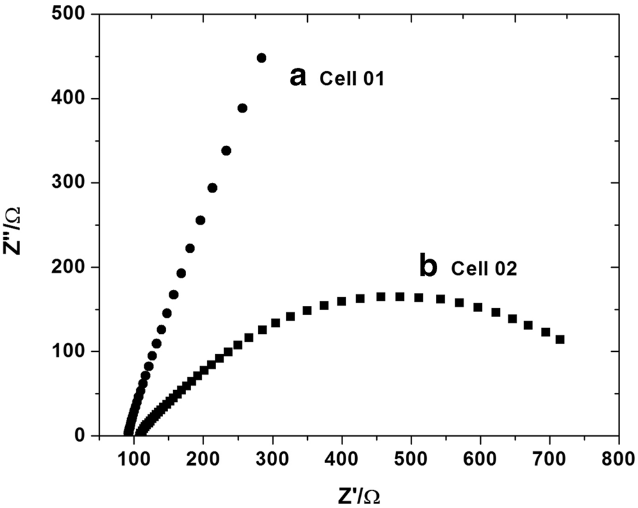

Fig. 4 Complex impedance plots for (*a*) cell 1: SS|[$(PEO)_{15}$:$Mg(Tf)_2$+ 10%PYR_{14}TFSI]|SS and (*b*) cell 2: Mg|[$(PEO)_{15}$:$Mg(Tf)_2$+ 10%PYR_{14}TFSI]|Mg recorded at room temperature

The oxidation of Mg to Mg^{2+} ions takes place during the discharge and the reverse process of electrodeposition of Mg takes place during charging. The voltammograms show the redox peaks corresponding to the deposition (−2.2 V) and dissolution of (2.0 V) of magnesium. Relatively high difference between anodic and cathodic peak potential value is due to the absence of a reference electrode in the system. Similar potential difference (~ 5 V) between Mg oxidation and reduction has been reported [11]. It shows that the charges associated with the deposition and dissolution process are nearly equal for the first cycle. This implies that, at the beginning, the reversibility of the magnesium deposition and dissolution is relatively high.

The cathodic and anodic peaks are distinctly observed for Mg electrolyte, where such feature is not observed for the cells with SS electrodes in the same potential range. This suggests that the cathodic deposition and anodic oxidation of Mg are facile at the Mg electrode and solid polymer electrolyte interface.

Charge/discharge characteristics of the fabricated cells were obtained by using a Metrohm Autolab potentiostat/galvanostat (PGSTAT 128N) system. Charge/discharge rates of 0.015 C (assuming that the theoretical capacity of TiO_2 is 330 mA hg^{-1}) were applied during Mg/[$(PEO)_{15}$:$Mg(Tf)_2$+ 10%IL]/TiO_2-C cell cycling within the 2.25–0.5 V potential window (Fig. 6).

Charge capacity of TiO_2 lies in the voltage range from 0.5 to 2.25 V, but during the discharge, most of capacity has

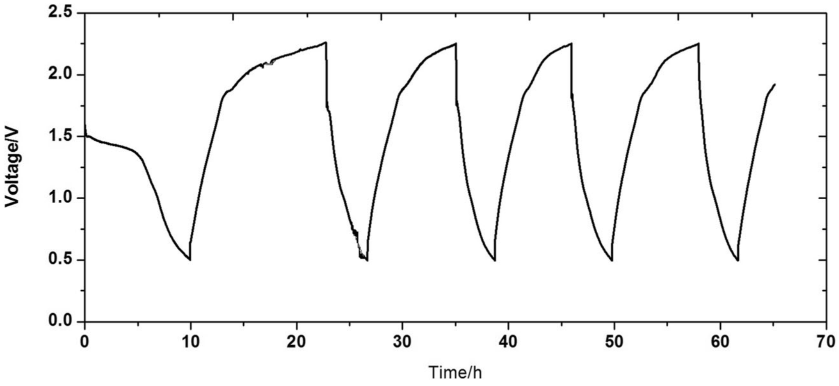

Fig. 6 Charge-discharge curves of a Mg/SPE/TiO_2-C cell at room temperature at 0.015 C rate

reduced below 1.75 V. In Fig. 6 during the discharge, sudden voltage drop can be observed from 2.25 V to just below 1.75 V. This voltage drop has not increased with the increase of the cycle numbers. Therefore, most of discharge capacity has lied below 1.75 V as shown in the voltage profile. Similar results with regard to sudden voltage drop have been reported by Narayan et al. [15] for their Mg^{2+} ion batteries with γ-MnO_2 and Mg metal as the electrodes together with the molten electrolyte, and the voltage drop has increased with the increase of cycle number. This is highly affected to the cyclability, durability, and the performance of the battery. According to the work reported in this paper, we observed that the sudden voltage drop during the cycling has not increased when the electrolyte incorporates with $PYR_{14}TFSI$ ionic liquid hence leading to enhance the overall battery performance. When the battery is subjected to continuous charge/discharge cycles, the first discharge (insertion of Mg^{2+} into the TiO_2 structure) yielded a capacity of 45 $mAhg^{-1}$ while the second discharge capacity is 22 $mAhg^{-1}$. Although a significant capacity fading has been observed for the first cycle, this fading has totally disappeared from the subsequent cycles.

Conclusion

A novel magnesium ion conduction solid polymer electrolyte of composition $[(PEO)_{15}:Mg(Tf)_2+10\%PYR_{14}TFSI]$ has been synthesized and characterized. An electrolyte of composition $[(PEO)_{15}:Mg(Tf)_2]$ without IL has also been prepared for comparison. On the basis of electrical and electrochemical studies, the following conclusions have been drawn. The solid polymer electrolyte film is flexible and offers relatively high ionic conductivity at room temperature. Mg^{2+} ion transport number has significantly increased by incorporating the 10 wt.% IL in to the IL-free electrolyte. Mg^{2+} ion transport number for $[(PEO)_{15}:Mg(Tf)_2+10\%PYR_{14}TFSI]$ electrolyte composition has the value of 0.4 while the electrolyte with composition $[(PEO)_{15}:Mg(Tf)_2]$ is 0.19. Studies based on AC impedance spectroscopy and cyclic voltammetry indicate the essential requirement of the electrochemical reversibility of Mg metal and Mg $^{2+}$ ions and hence confirm the Mg ion conduction in the solid polymer electrolyte. Rechargeable Mg^{2+} ion batteries were fabricated with low-cost TiO_2 as the active cathode material, a PEO-based solid state polymer membrane as the electrolyte, and Mg metal as the anode. Preliminary studies on rechargeable batteries Mg/$[(PEO)_{15}:Mg(Tf)_2+10\%IL]/TiO_2$-C show 45 $mAhg^{-1}$ discharge capacity.

References

1. Tarascon JM, Armand M (2001) Issues and challenges facing rechargeable lithium batteries. Nature 414:359–367
2. Kumar GG, Munichandraiah N (1999) Reversibility of Mg/Mg 2+ couple in a gel polymer electrolyte. Electrochim Acta 44:2663–2666
3. Connor JH, Reid WE, Wood GB (1957) Electrodeposition of metals from organic solutions V. Electrodeposition of magnesium and magnesium alloys. J Electrochem Soc 104:38–41
4. Cheek G, O'Grady W, El Abedin SZ, Moustafa E, Endres F (2008) Studies on the electrodeposition of magnesium in ionic liquids. J Electrochem Soc 155:D91–DD5
5. Inamoto M, Kurihara H, Yajima T (2013) Vanadium pentoxide-based composite synthesized using microwave water plasma for cathode material in rechargeable magnesium batteries. Materials 6:4514–4522
6. Pandey G, Hashmi S (2009) Experimental investigations of an ionic-liquid-based, magnesium ion conducting, polymer gel electrolyte. J Power Sources 187:627–634
7. Yoshimoto N, Shirai T, Morita M (2005) A novel polymeric gel electrolyte systems containing magnesium salt with ionic liquid Electrochimica acta 50: 3866–3871
8. Kim JK, Matic A, Ahn JH, Jacobsson P (2010) An imidazolium based ionic liquid electrolyte for lithium batteries. J Power Sources 195:7639–7643
9. Armand M, Endres F, MacFarlan DR, Ohno H, Scrosati B (2009) Ionic-liquid materials for the electrochemical challenges of the future. Nat Mater 8:621–629
10. Pitawala J, Navarra MA, Scrosati B, Jacobsson P, Matic A (2014) Structure and properties of Li-ion conducting polymer gel electrolytes based on ionic liquids of the pyrrolidinium cation and the bis (trifluoromethanesulfonyl) imide anion. J Power Sources 245:830–835
11. Kumar Y, Hashmi S, Pandey G (2011) Ionic liquid mediated magnesium ion conduction in poly (ethylene oxide) based polymer electrolyte. Electrochim Acta 56:3864–3873

12. Evans J, Vincent CA, Bruce PG (1987) Electrochemical measurement of transference numbers in polymer electrolytes. Polymer 28: 2324–2328
13. Kumar GG, Munichandraiah N (2002) Poly (methylmethacrylate)—magnesium triflate gel polymer electrolyte for solid state magnesium battery application. Electrochim Acta 47:1013–1022
14. Bruce P, Vincent C (1993) Polymer electrolytes. J Chem Soc Faraday Trans 89:3187–3203
15. Narayanan NV, Raj BA, Sampat S (2009) Magnesium ion conducting, room temperature molten electrolytes. Electrochem Commun 11:2027–2031