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# Ionic Liquid-Based Gel Polymer Electrolytes for Application in Rechargeable Lithium Batteries

*Himani Gupta and Rajendra K. Singh*

## Abstract

Depleting fossil fuels has put pressing need for the search of alternative energy resources. Solar and wind energy resources are being considered one of the viable solutions. However, these intermittent sources require efficient energy storage systems in terms of rechargeable Li batteries. In Li batteries, electrolyte is one of the most important components to determine the performance, as it conducts the ions between the electrodes. In battery, mostly liquid electrolyte is used as it shows high ionic conductivity and electrode/electrolyte contact which help to reduce the internal resistance. But these are not electrochemically very stable and raised some major problems such as reactivity with electrode, dissolution of electrode ions, leakage, volatility, fast Li dendrite growth, etc. Therefore, in order to improve its electrochemical performance, selection of electrolyte is an important issue. In the present study, ionic liquid (IL)-based polymer electrolyte is used over liquid electrolyte in which IL acts as a plasticizer and improves ionic conductivity and amorphicity. These electrolytes have high thermal and electrochemical stability, therefore, can be used in high voltage Li battery. Also, their mechanical stability helps to suppress Li dendrites growth. Therefore, polymer electrolytes can open a new way in the progression of battery application.

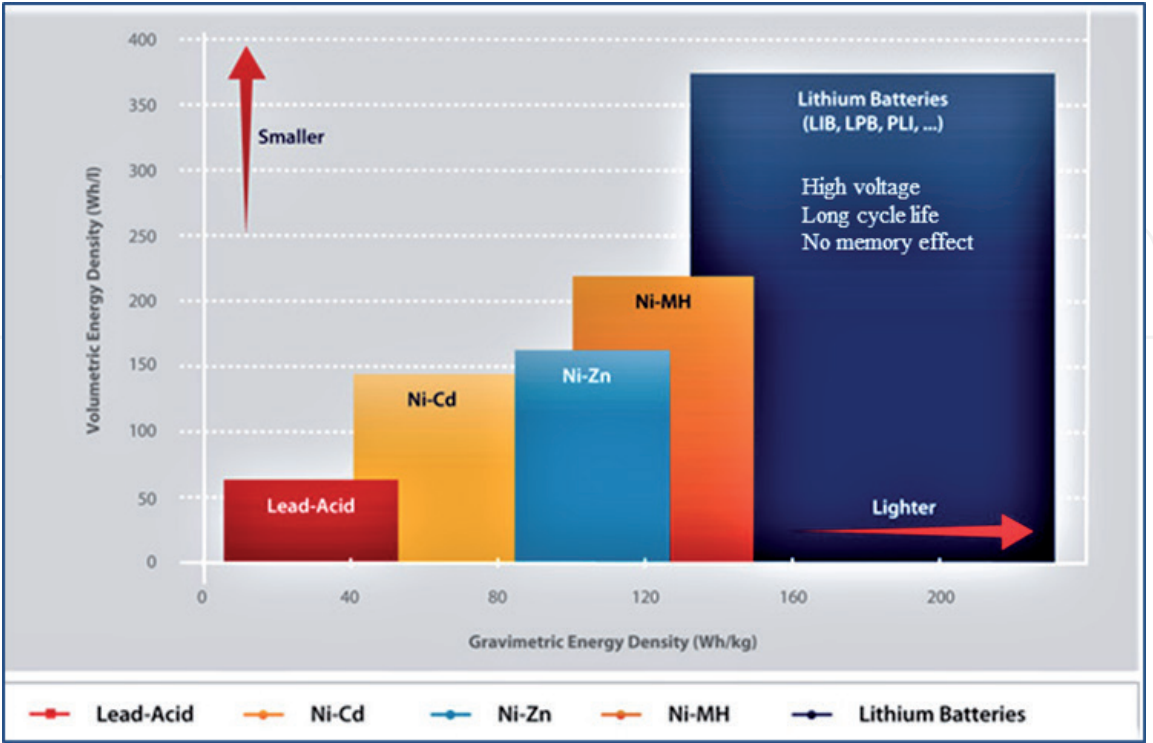
**Keywords:** ionic liquid, polymer electrolyte, ionic conductivity, electrochemical analysis, Li battery

## 1. Introduction

Energy contributes a major role in various aspects of human life and their demand is continuously increasing with time. To meet this challenge, world population mostly depends on fossil fuels which include petroleum, coal, nuclear power and natural gases. But the climate change due to the frequently increasing CO<sub>2</sub> level has driven the research in the development of renewable energy sources. Research in the field of renewable energy sources such as solar energy, wind energy and hydropower has been initiated but the major problem to efficiently utilize these energies is to investigate suitable electrical energy storage devices because these energy sources are intermittent in nature. For this purpose, the most efficient energy storage devices are the batteries and super capacitors [1, 2]. Both the devices have their own importance depending on usage but batteries are continuing to dominate in the market of portable electronic system because of their

high energy density and voltage rating [3]. Battery is an electrochemical device which can convert chemical energy into electrical energy through redox reaction to release the energy. Conversely, it can also convert electrical energy into chemical energy to store the energy. Among all the batteries, rechargeable lithium batteries (Li-batteries) are gaining much attention in the electric power storage system due to their high capacity, working voltage, long lifetime, low self-discharge rate and no memory effect (**Figure 1**) [4, 5]. However, the energy density of recent Li batteries is in the range of 100–200 Wh/Kg which limits their use in automotive application [6]. Therefore, to enhance their energy density, Li metal is frequently used because it shows very high capacity for Li battery. Metallic lithium as anode is being used since long time but its application with organic liquid electrolytes arises the main issue in lithium batteries as lithium dendrite growth [7–9]. Further, the use of these volatile and flammable electrolytes causes safety problems during cycling. Also, these organic liquid electrolytes cannot be used in high voltage batteries due to their electrochemical instability at higher voltage [10].

Therefore, for safely utilization of Li metal in batteries, requirement of alternative electrolyte is highly demanded. In this context, polymer electrolytes are gaining much attention in Li batteries because of their outstanding properties such as mechanical, thermal and electrochemical stability, safety and flexibility [11, 12]. The polymer acts as a host matrix for ion movement in which ions can move in the free space provided by the polymer matrix. Generally, solid polymer electrolytes (SPEs) are formed by dissolving organic salt into the polymer matrix. The selection of polymer matrix mainly depends on the presence of polar group so that it can easily coordinate with the cations; and there is less restraint in the bond rotation [13]. Among the different polymer matrices, poly (ethylene oxide) (PEO) based polymer electrolytes are mostly studied due to its high chain flexibility and ability to dissolve different organic/inorganic materials [14, 15]. However, PEO based solid electrolytes are semi-crystalline in nature which comprise the crystalline and

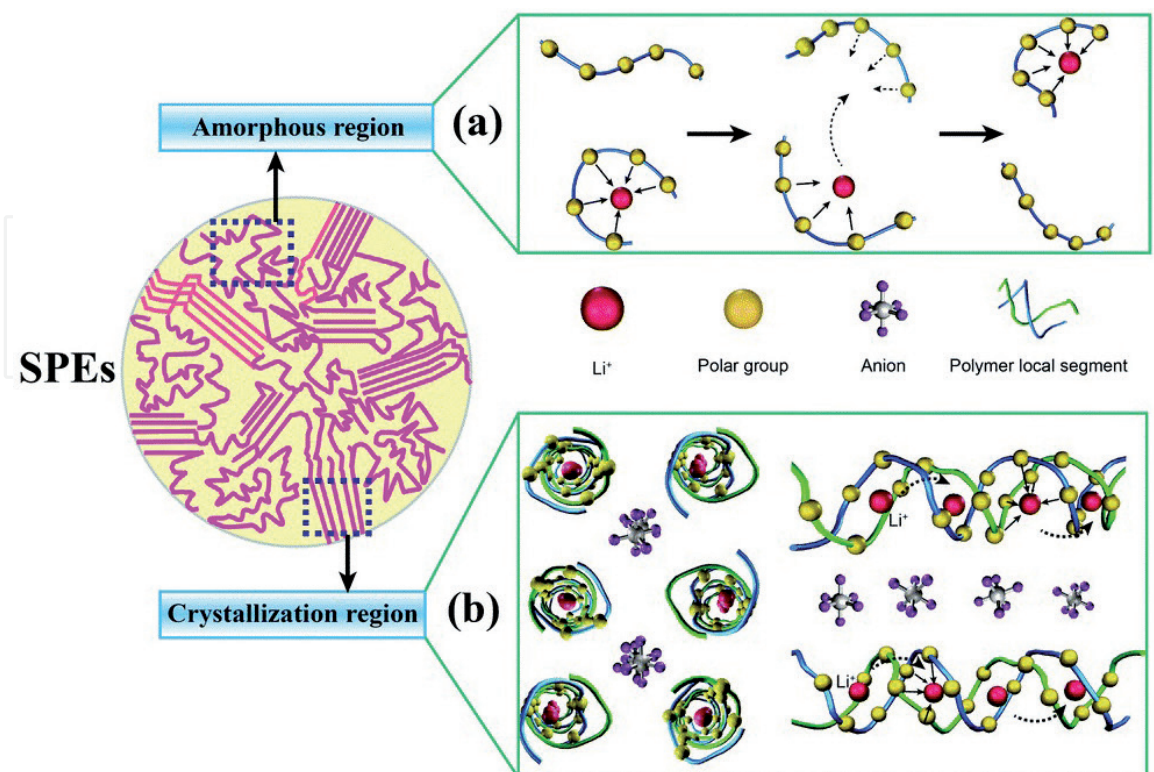


**Figure 1.**  
Comparison in different types of batteries.

amorphous regions both; and it is reported that the ion conduction in polymer electrolytes occurs only in amorphous region [16–18]. Therefore, they show lower room temperature ionic conductivity ( $10^{-8}$  to  $10^{-6}$  S/cm) and hence cannot be used in practical battery application. To overcome this drawback and to obtain polymer electrolytes having good ionic conductivity, flexibility, mechanical and electrochemical stability, different approaches are reported such as the addition of nanofillers, use of plasticizers or gel polymer electrolytes (GPEs) [19, 20]. Among these approaches, use of GPEs is in focus as they combine the desirable properties of both liquid (high conductivity) as well as solid (mechanical stability). Thus it is a suitable replacement of electrolyte for high performance batteries. In the present chapter, fundamental properties of PEO based polymer electrolytes; their classifications and performance in Li batteries are discussed.

## 2. Properties and ion transport mechanism in PEO

PEO has chemical structure  $\text{H}-(\text{O}-\text{CH}_2-\text{CH}_2)_n-\text{OH}$  which is a polyether compound. Depending on the molecular weight of polymer, it is also known as polyethylene glycol (PEG). Generally, polymer having molecular weight above 20,000 g/mol is called PEO while, below 20,000 g/mol is known as PEG [21]. PEO is a low toxic compound; therefore, it is widely used in many applications such as chemical, industrial, medical, biological etc. PEO has ethylene oxide unit which provides flexibility and high donor number for  $\text{Li}^+$  ions and promotes the ion transport. Also, it has high dielectric constant and  $\text{Li}^+$  ion solvating ability [22]. The ion transport mechanism in PEO occurs by the coordination of  $\text{Li}^+$  ions with the ether oxygen atom of PEO chain. This process occurs by formation and dissociation of  $\text{Li}-\text{O}$  bond by local segmental motion of polymer chain (Figure 2) [23]. The ion transportation in polymer



**Figure 2.**  
 Ion transport mechanism in polymer PEO in (a) amorphous as well as (b) crystalline region.



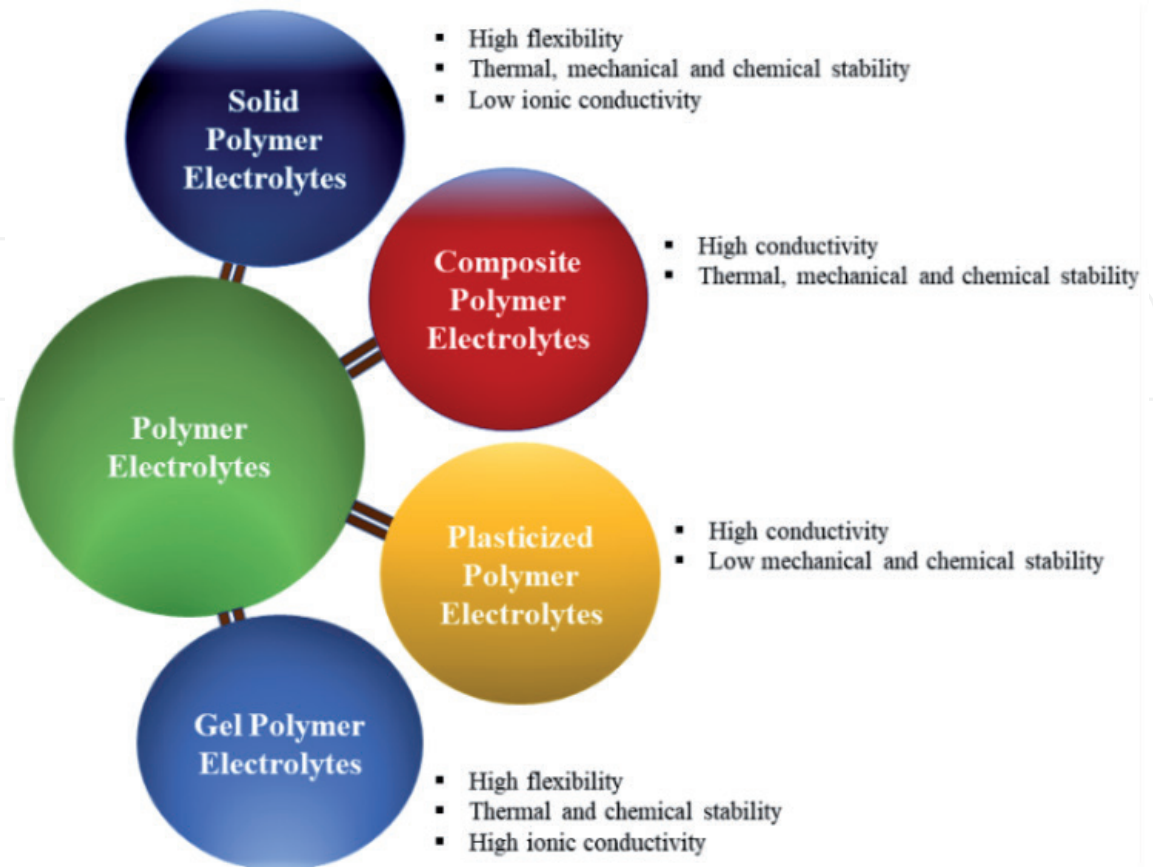
electrolytes mainly depends on polymer chain segmental motion which becomes faster in amorphous region or above the glass transition temperature ( $T_g$ ) of polymer (as shown in **Figure 2(a)**). Hence, ideally polymer should have lower  $T_g$  to remain in rubbery state or to promote the  $\text{Li}^+$  ion conductivity at RT [17]. In order to enhance  $\text{Li}^+$  ion conductivity of polymer electrolytes different strategies such as addition of nano fillers, use of plasticizers or gel polymer electrolytes has been proposed. On this basis, polymer electrolytes are classified into different categories as discussed below.

### 3. Classification of polymer electrolytes

Polymer electrolytes are considered to be promising materials in the research and development of electrochemical devices. On the basis of materials, polymer electrolyte is classified into following categories (**Figure 3**):

#### 3.1 Dry solid polymer electrolytes

It is formed by incorporating inorganic salt into the polar polymer thus ion conducting electrolyte is known as solid polymer electrolyte [24]. The electrostatic interaction between the metal ions of salt and polar polymer results the formation of coordination bond. This metal-polymer interaction can be affected by many factors such as nature and distance between the functional group of polymer, molecular weight, nature of branching, charge on metal and counter ion [25]. When the polymer electrolyte is placed in the electric field, ions start to move from one coordination site to other. It occurs due to weaker interaction between the metal ion and functional group of polymer chain.



**Figure 3.**  
*Differently types of polymer electrolytes used in Li battery.*

### 3.2 Plasticized polymer electrolytes

Plasticized polymer electrolytes are formed by dissolving low molecular weight compounds (e.g. ethylene carbonate (EC), propylene carbonate (PC), poly ethylene glycol (PEG)) [26]. These plasticizers reduce the inter and intra-molecular interaction between the polymer chain, thus reduce the  $T_g$  and crystallinity of polymer chain and enhance the salt dissociation ability [27]. Although this approach improves the conductivity of polymer electrolyte, but it also provides low mechanical stability, solvent volatility and reactivity with lithium electrode.

### 3.3 Composite polymer electrolytes

Conductivity of polymer electrolytes also decreases due to the presence of ion pair formation. This behavior is observed mainly because of the low dielectric constant of polymer matrix [28]. To overcome this issue, high dielectric constant inorganic inert fillers (such as  $TiO_2$ ,  $SiO_2$ , etc.) are dispersed into the polymer electrolyte to avoid ion-ion association. The obtained electrolyte combines the properties of ceramic fillers as well as polymer and results the flexible, mechanically stable composite polymer electrolyte [29].

### 3.4 Gel polymer electrolytes

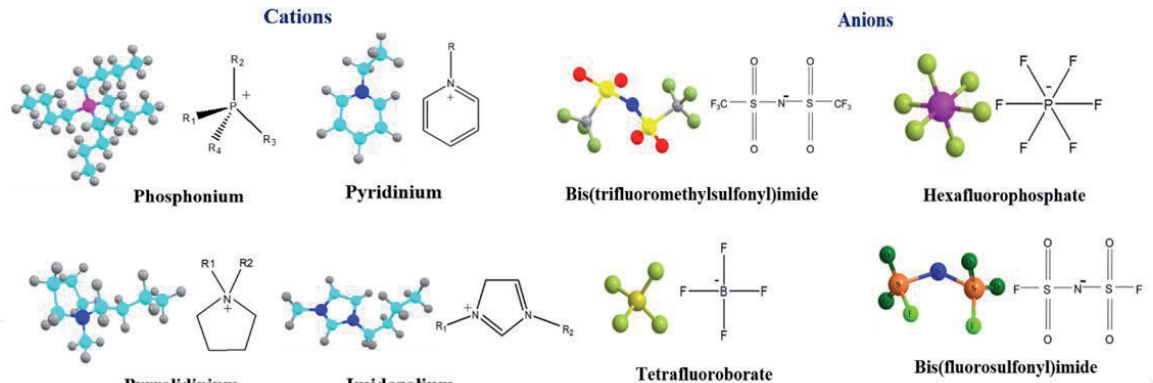
In all the polymer based electrolytes, gel polymer electrolytes (GPEs) are focusing much attention because they combine the advantages of liquid electrolyte such as high conductivity, good electrode/electrolyte contact and solid electrolytes like as safety, mechanical and thermal stability. In GPEs, polymer is used to trap the liquid constituent and provides mechanical support, thus it is considered as safer than liquid electrolytes. For the synthesis of GPEs, large amount of organic solvent is used in the polymer in which its conduction takes place along the host polymer [30]. Recently, ionic liquid (IL) based GPEs are in focus of the research due to the desirable properties of ILs such as high conductivity, thermal stability, negligible vapor pressure which makes GPE as a suitable replacement of liquid electrolyte.

## 4. Ionic liquid

ILs are the molten salts that remain in liquid state below  $100^\circ\text{C}$ . Sometimes these are also referred as room temperature molten salts, ionic fluids, fused salts or organic salts. ILs are generally formed by self-dissociated, poorly coordinated, bulky organic cations and organic/inorganic anions [31]. Some of the common cations and anions of ILs are given in **Figure 4**. These ILs do not have strong ionic bond between the cations and anions as in ionic salts ( $NaCl$ ,  $KCl$ , etc.), hence possess low lattice energy and remain in dissociated state. Therefore, they show many desirable properties as high conductivity, low vapor pressure, melting and glass transition temperature, high thermal and electrochemical stability, less polluting and easily recyclable [32]. Some of the properties of ILs are given in **Table 1** [33–35].

### 4.1 IL based gel polymer electrolytes

ILs provide outstanding ionic conductivity upto the decomposition temperature which enable them to be used as electrolyte. Also, to eliminate their leakage issue, ILs are trapped in polymer matrices which are known as GPEs. These IL based GPEs show high conductivity, good thermal and electrochemical stability, transparency and flexibility (**Figure 5**) [36].



ILs based GPEs	Conductivity (mS/cm)	References
PEO <sub>20</sub> LiTFSI <sub>2</sub> [Pyr <sub>1,20</sub> TFSI] <sub>4</sub>	2.5 × 10 <sup>-4</sup> at 20°C	[37]
PEO <sub>20</sub> LiTFSI <sub>1</sub> [Pyr <sub>1,20</sub> TFSI] <sub>1.5</sub>	7 × 10 <sup>-5</sup> at 20°C	[35]
PVdF-HFP + 20 wt.% LiTFSI+60% BMIMBF <sub>4</sub>	1.7 at RT	[38]
PPEGDA <sub>15</sub> %[LiBF <sub>4</sub> Im <sub>12</sub> BF <sub>4</sub> ] <sub>85</sub> %	1.2 × 10 <sup>-4</sup> at 20°C	[39]
[PPyr <sub>11</sub> TFSI] <sub>50</sub> %[Li(G <sub>4</sub> )]TFSI <sub>50</sub> %	1 × 10 <sup>-4</sup> at 20°C	[40]
EGDMA-MMA + 0.5 M PP <sub>14</sub> Cl + 80% PP <sub>14</sub> TFSI	0.09 at 25°C	[41]
PEO + 20wt%LiFSI+7.5wt%EMIMFSI	0.289 at RT	[36]
PEO + 20wt%LiTFSI+12.5wt%EMIMTFSI	0.208 at 30°C	[42]
PEO <sub>20</sub> LiTFSI [Pyr <sub>13</sub> TFSI] <sub>2,15</sub>	3 × 10 <sup>-4</sup> at 20°C	[43]
PEO + 10wt%NaMS+60wt%BMIM-MS	0.105 at 30°C	[44]
PEO + 10% NaTFSI+40% BMIMTFSI	0.4 at 30°C	[45]
PEO <sub>20</sub> LiTFSI <sub>2</sub> [Pyr <sub>14</sub> TFSI] <sub>2</sub>	1 × 10 <sup>-4</sup> at 20°C	[46]

**Table 2.**  
 Ionic conductivity of ILs based GPEs.

5. Results and discussion

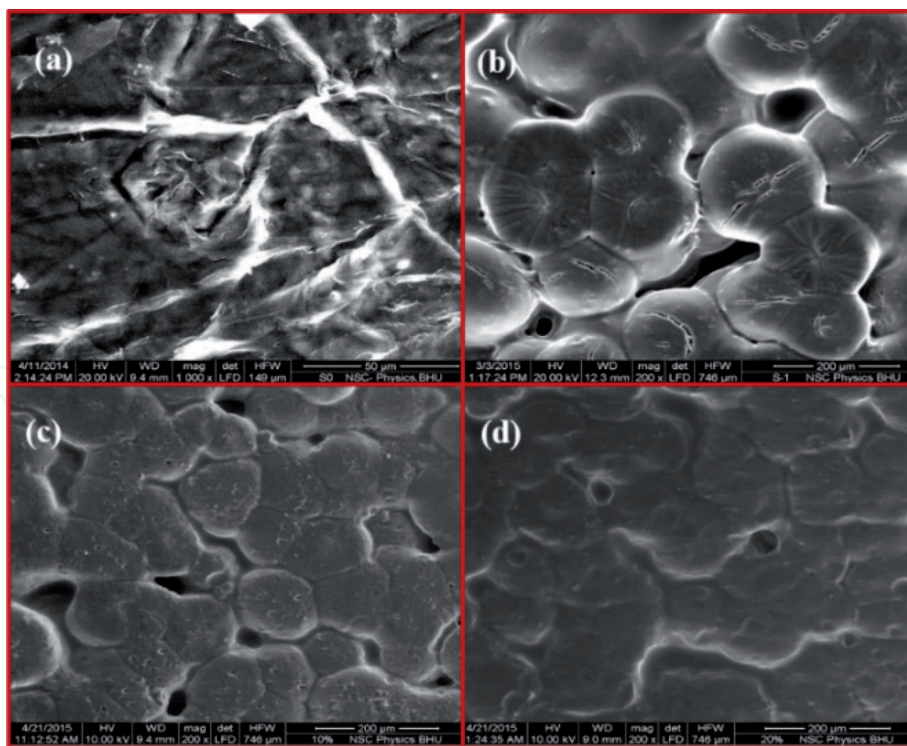
5.1 Surface morphology and crystallinity of IL based GPEs

The effect on surface morphology and degree of crystallinity of PEO based SPE with the addition of IL is reported in literature. Gupta et al. [47] studied that, by using the phosphonium based IL (Trihexyltetradecylphosphonium TFSI) in polymer electrolyte (PEO + 20 wt% LiTFSI), increment in amorphous region was obtained (**Figure 6**). The surface morphology of PEO based GPE is given in **Figure 6**. **Figure 6(a)** shows the crystalline region of polymer PEO, when IL (up to 20 wt%) was incorporated into PEO-LiTFSI system, smoother morphology was observed which resulted the amorphous nature of GPE. Singh et al. [48] also reported that the suppression in rough surface nature of polymer electrolyte (PEO + 20 wt% LiTFSI) was observed when BMIMTFSI IL was added into it. The smoother surface morphology of polymer electrolyte was observed due to the plasticization effect of ionic liquid which reduced the interaction between the polymer chain and made it more flexible.

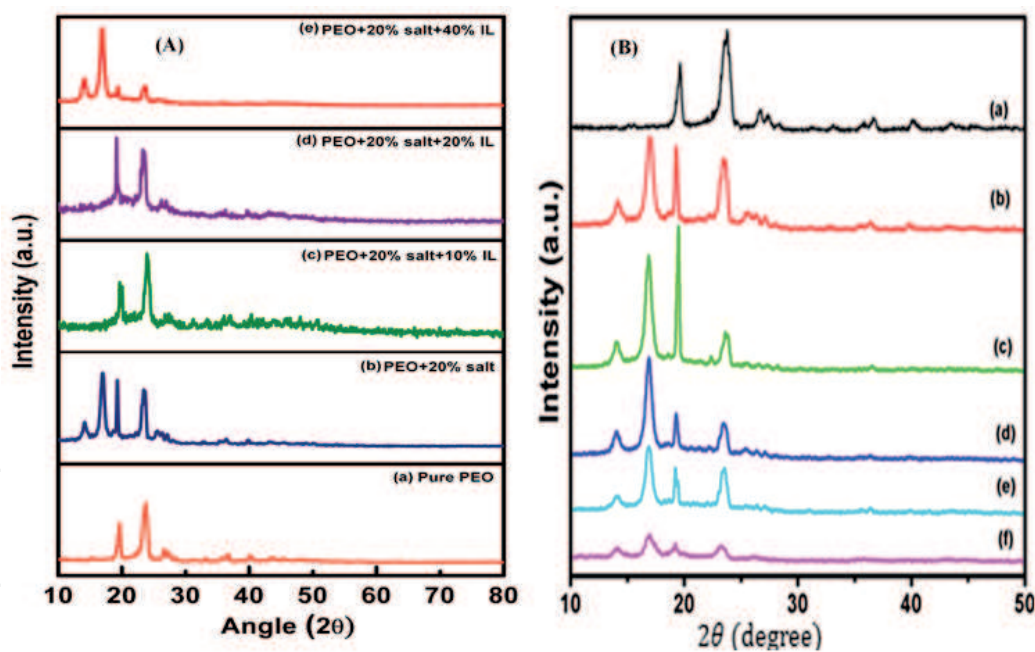
From the XRD analysis, crystallinity of GPEs is also reported in many studies. Gupta et al. [47] reported the variation in crystallinity of SPE with the addition of phosphonium based IL They showed the semi-crystalline nature of polymer PEO. When LiTFSI salt and IL were added into the PEO, broadening of halo region and FDHM of polymer electrolytes were noticed which resulted the decrement in crystalline region or enhancement in amorphous region of polymer electrolyte (**Figure 7(A)**).

Singh et al. [48] reported the effect of BMIMTFSI IL on the crystallinity of SPE (PEO + 20 wt% LiTFSI). They showed that with the addition of IL into SPE, halo region was increased substantially and relative intensities of the crystalline peaks reduced. This proved the enhancement in amorphous phase in polymer electrolyte (**Figure 7(B)**). Therefore, presence of IL in polymer electrolyte improves its amorphous region which is desirable for the conduction of Li<sup>+</sup> ions, since, conduction in polymer electrolytes occurs only in this region.





**Figure 6.**  
SEM image for (a) pure PEO and PEO + 20 wt% LiTFSI +  $X$ wt% IL (b)  $X = 0$ , (c)  $X = 10$ , (d)  $X = 20$ .



**Figure 7.**  
XRD pattern of (A) phosphonium IL based polymer electrolytes, PEO + 20wt.%LiTFSI +  $x$ % IL ( $x = 0, 10, 20, 40$ ); and (B) polymer electrolytes (a) PEO with PEO + 20wt.%LiTFSI +  $x$ wt.% BMIMTFSI (b)  $x = 0$ , (c)  $x = 5$ , (d)  $x = 10$ , (e)  $x = 15$  and (f)  $x = 20$ .

## 5.2 Ionic and $\text{Li}^+$ ions conductivity of IL based GPEs

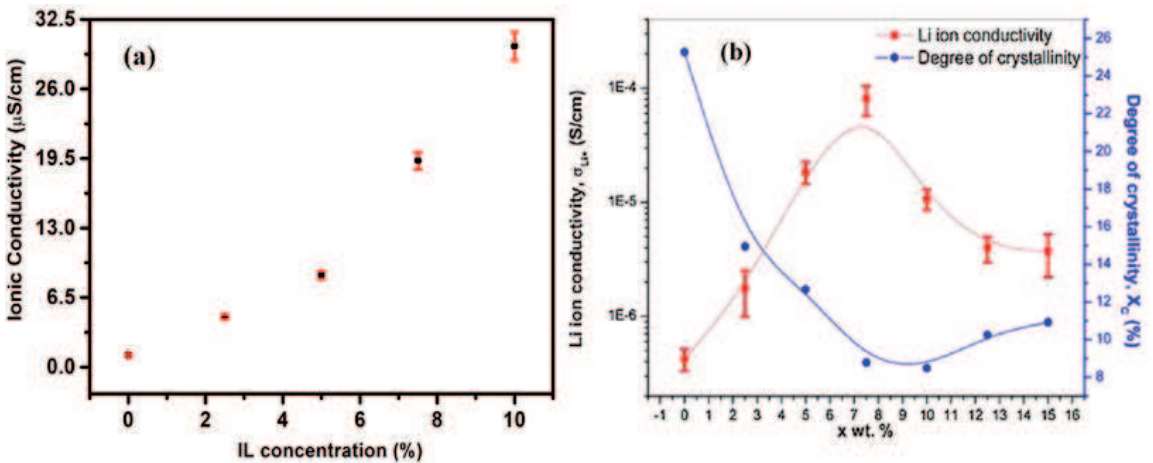
In batteries, electrolyte plays the major role to transport the ions between the two electrodes. So, conductivity of electrolyte is very important parameter for Li battery. It is well known that the IL acts as a plasticizer and its presence in polymer electrolyte enhances the ionic conductivity. IL also provides free charge carriers and therefore helps to promote the ionic conductivity of polymer electrolyte.

The increment in the conductivity of polymer electrolyte with IL concentration is also reported by Gupta et al. [49] and Balo et al. [36] which are depicted in **Figure 8(a)** and **(b)** respectively. Gupta et al. [49] showed that ionic conductivity of polymer electrolyte (PEO + 20 wt% LiFSI) increases with IL (PYR<sub>13</sub>FSI) concentration.

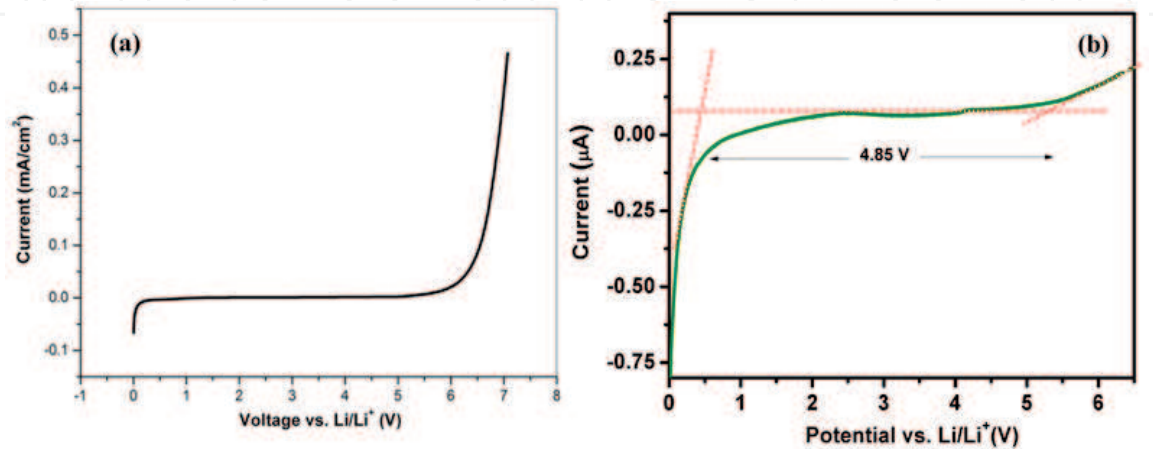
Whereas, Balo et al. [36] synthesized the PEO based polymer electrolyte (PEO + 20wt.%LiFSI + xwt.%EMIMFSI; for 0 ≤ x ≤ 15) and mentioned the use of optimized concentration of IL in polymer electrolyte. They found similar increasing trend of conductivity of polymer electrolyte with IL, but after certain concentration, it showed decreasing trend (**Figure 8(b)**). It happened because with the addition of IL into polymer electrolyte, large number of FSI<sup>-</sup> anions was available which started to interact with Li<sup>+</sup> ions present in the polymer electrolyte. Therefore, they formed ion pairs instead of participating in interaction with ether oxygen of PEO, due to which conductivity of electrolyte was reduced.

### 5.3 Electrochemical stability of IL based GPEs

Electrochemical stability of electrolyte is an important parameter as it decides the performance of the battery in working voltage range. Electrochemical stability of GPEs should be high so that it can be used in high voltage Li batteries. Many studies have been carried out on the electrochemical performance of PEO and IL



**Figure 8.** Variation in (a) ionic conductivity as well as (b) Li<sup>+</sup> ion conductivity and degree of crystallinity of polymer electrolyte with IL concentration.



**Figure 9.** Electrochemical stability of (a) imidazolium (EMIMFSI) and (b) pyrrolidinium (PYR<sub>13</sub>FSI) IL based GPEs.

based GPEs. Singh et al. [48] synthesized the GPE, PEO + 20 wt% LiTFSI + xwt% BMIMTFSI ( $x = 5, 10, 15, 20$ ) for Li battery and the electrochemical stability of 20 wt% IL containing GPE was reported  $\sim 4$  V vs. Li/Li<sup>+</sup>.

Balo et al. used the same polymer system with EMIMFSI and EMIMTFSI ILs and found that the GPE, PEO + 20 wt% LiTFSI + 10 wt% EMIMFSI was electrochemically stable up to  $\sim 6.4$  V vs. Li/Li<sup>+</sup> (**Figure 9(a)**) [50]. In another study they showed that the polymer electrolyte, PEO + 20 wt% LiTFSI + 10 wt% EMIMTFSI, was stable upto  $\sim 5.1$  V vs. Li/Li<sup>+</sup> [42]. Also, Gupta et al. [49] reported the electrochemical stability of pyrrolidinium IL based GPE (PEO + 20% LiFSI + 10% PYR<sub>13</sub>FSI) which was  $\sim 4.8$  V vs. Li/Li<sup>+</sup> (**Figure 9(b)**). Therefore, it can be concluded that IL based GPEs show enough electrochemical stability to be used in high voltage Li batteries application.

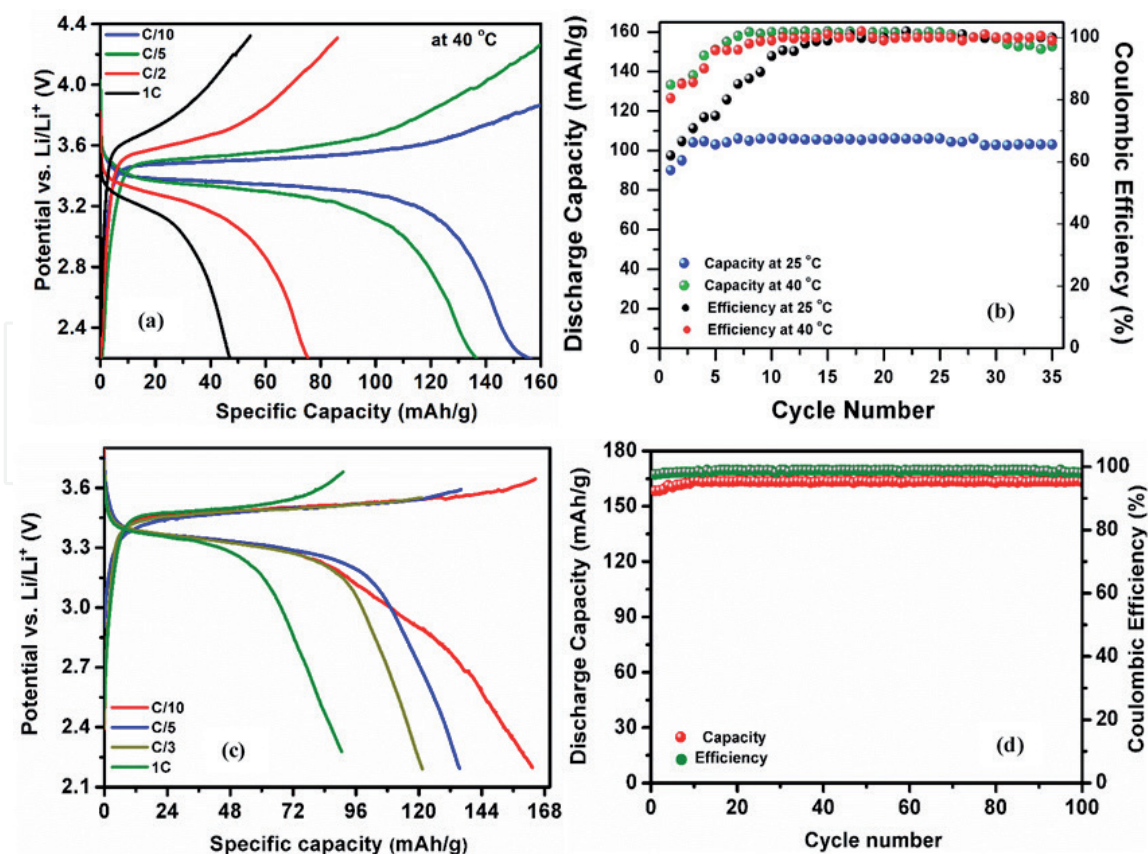
## 6. Application of IL based GPEs in Lithium batteries

Because of having several required properties, IL based polymer electrolytes are frequently used in many application as in supercapacitors, batteries and fuel cell etc. Particularly, in Li batteries, they are gaining much attention due to their high energy density, flexibility and safety. In recent years, almost all the electronic equipment are being run by polymer batteries as laptop, mobile phones, power banks, portable media players etc. Many reports include the application of GPEs in Li batteries. As one of the main advantage of GPE is that it forms stable solid electrolyte interface (SEI) passive layer between the electrode-electrolyte and provides higher cyclic stability to Li battery. Battery performance of PEO based GPEs have already been studied in literature. Gupta et al. [51] synthesized the GPE, (PEO + 20 wt% LiTFSI + 30 wt% 1-butyl-3-methyl pyridinium TFSI), and reported its performance in Li battery in (Li/LiFePO<sub>4</sub>) configuration. They obtained maximum discharge capacity  $\sim 160$  mAh/g and 99% Coulombic efficiency upto 35 cycles at 40°C (**Figure 10(a)** and **(b)**). In another report, they used the pyrrolidinium based IL in polymer system, PEO + 20% LiFSI + 10% PYR<sub>13</sub>FSI, with graphene oxide coated LiFePO<sub>4</sub> cathode and obtained maximum discharge capacity  $\sim 163$  mAh/g at C/10 rate at room temperature (RT) (**Figure 10(c)**) [49]. It was the result of high conductivity of IL, PYR<sub>13</sub>FSI and LiFSI salt as well as high electronic conductivity and large surface area of graphene oxide (GO) which enhanced the electron transfer rate and hence capacity of Li battery. This Li battery provided almost constant capacity and Coulombic efficiency upto 100 cycles (**Figure 10(d)**). Balo et al. [36] reported the same system (Li/LiFePO<sub>4</sub>) using imidazolium IL based polymer electrolyte PEO + 20 wt.% LiFSI + 7.5wt.%EMIMFSI at RT.

They found maximum discharge capacity  $\sim 143$  mAh/g at C/20 rate which decreased upto 130 mAh/g at C/10 and further reduced upto 20 mAh/g at 2C discharge rate. This reduction of discharge capacity was reported due to the increase of electrolyte ohmic drop and limited Li<sup>+</sup> ion diffusion in composite cathode. The above polymer systems have been also tested with high voltage and capacity cathode materials such as LiNi<sub>x</sub>Mn<sub>y</sub>Co<sub>z</sub>O<sub>2</sub> (NMC) and LiNi<sub>x</sub>Co<sub>y</sub>Al<sub>z</sub>O<sub>2</sub> (NCA). These electrolytes are electrochemically stable even at high voltage which deliver high capacity and cyclic stability to the Li battery. Gupta et al. [52] used the phosphonium based IL (trihexyltetradecylphosphonium bis TFSI) in PEO-LiTFSI polymer system.

They fabricated the Li cell (Li/LiNi<sub>0.6</sub>Mn<sub>0.2</sub>Co<sub>0.2</sub>O<sub>2</sub>) and obtained maximum discharge capacity  $\sim 148$  mAh/g with 95% Coulombic efficiency upto 150th cycle at C/10 rate in the voltage range of 2.4–4.2 V (**Figure 11(a, b)**). The impedance of the Li cell was also evaluated with cycling (inset of **Figure 11(b)**). It showed the slight increment in the interfacial resistance value and hence, resulted very small capacity fading of Li cell (**Figure 11(b)**). Balo et al. [50] used imidazolium IL (EMIMFSI)

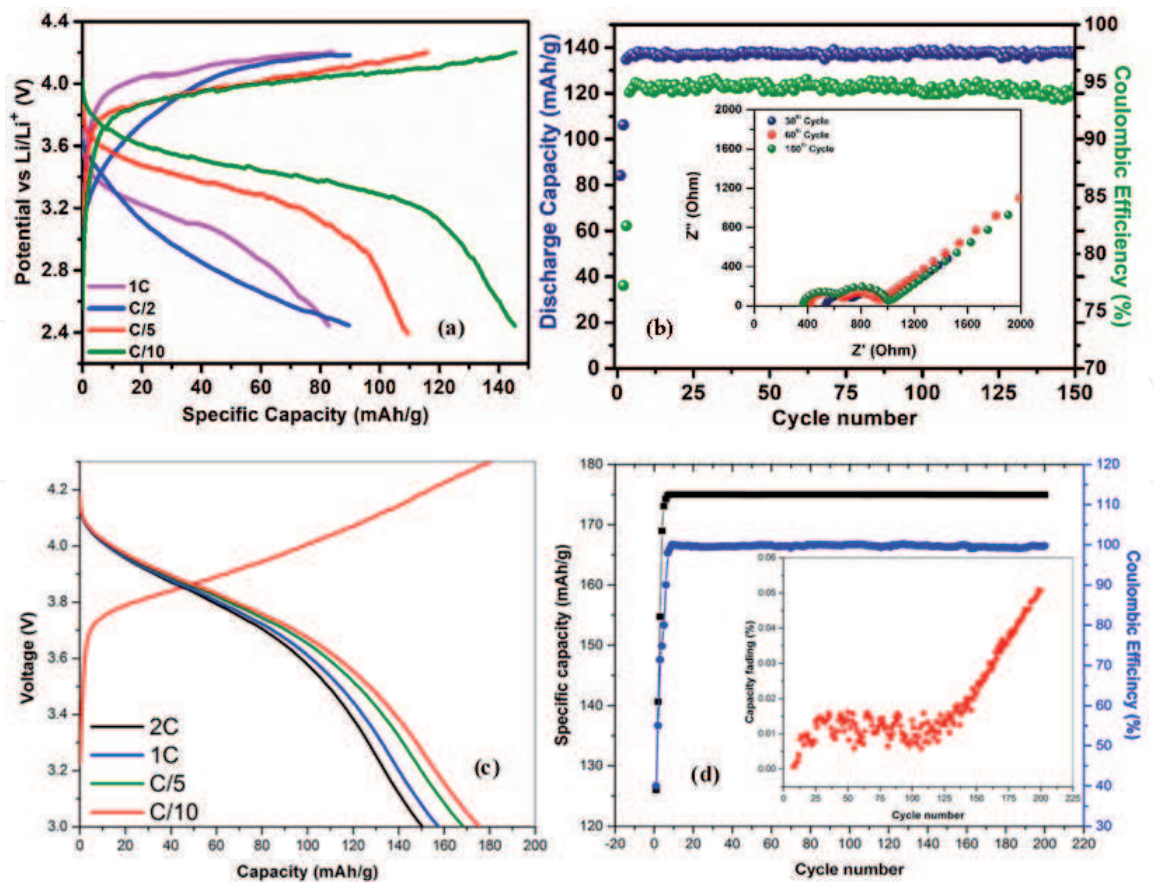




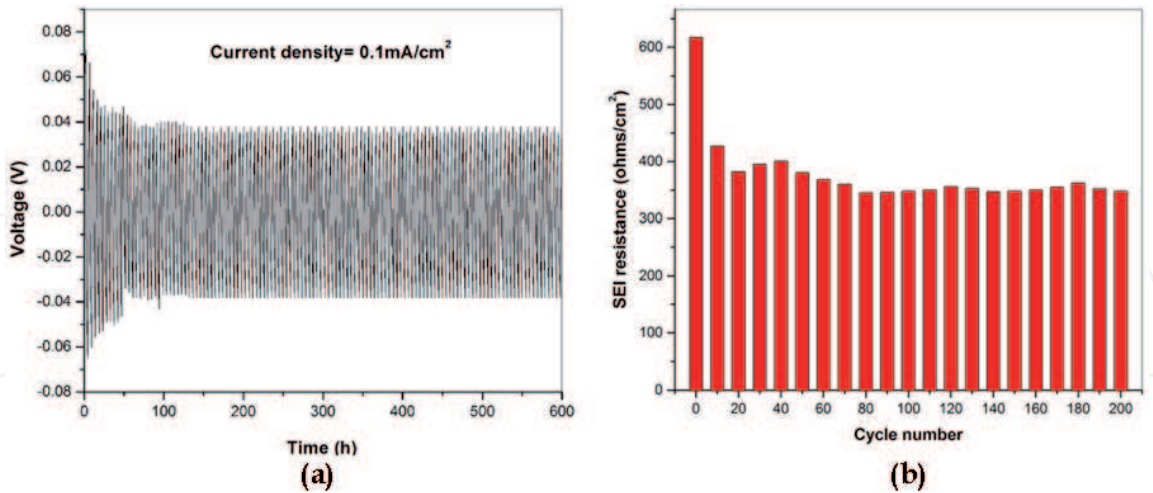
**Figure 10.**  
 Electrochemical performance of (a, b) 1-butyl-3-methyl pyridinium TFSI and (c, d) N-propyl-N-methylpyrrolidinium-FSI IL based polymer electrolytes with  $\text{Li/LiFePO}_4$  and graphene oxide coated  $\text{LiFePO}_4$  cathode respectively.

based GPE (PEO + 20 wt% LiTFSI + 10 wt% EMIMFSI) in Li/NCA cell. They observed the discharge capacity  $\sim 175$  mAh/g at C/10 current rate which remained almost stable throughout cycling (**Figure 11(c, d)**) and only 0.05% of total capacity was lost during 200 cycles (inset of **Figure 11(d)**). The use of Li metal electrode in Li batteries are in demand due to its higher energy density and capacity (3862 mAh/g), but it could not be frequently used in application purpose because of the formation of Li dendrites. This Li dendrite is formed due to the deposition of  $\text{Li}^+$  ions on the Li metal surface during cycling which starts to grow and causes short circuiting and results low cyclic stability. Therefore, in order to obtain high capacity and safer Li battery, suppression of dendrite growth is important. It was reported that the dendrite growth becomes faster with liquid solvents. But the use of GPEs in Li battery is able to suppress its growth because of having mechanical stability. Therefore, the use of GPEs provides safety and cyclic stability to Li battery. The electrochemical stability of GPEs with Li electrode is reported in literature. Wang et al. [53] reported the combination of the use of LAGP-PEO (LiTFSI) composite solid electrolyte and the modification of Li anode with  $\text{PEO}_{500000}$  (LiTFSI) in  $\text{Li/LiMn}_{0.8}\text{Fe}_{0.2}\text{PO}_4$  battery. They obtained that the use of both can effectively prevent the Li dendrite growth. Kim et al. used three different ILs (BMITFSI, BMIBF<sub>4</sub> and BMICF<sub>3</sub>SO<sub>3</sub>) in polymer system (PEO-LiTFSI) and reported that the Polymer electrolyte with BMITFSI IL resulted low and stable interfacial resistance or dendrite growth on lithium metal. Balo et al. [50] examined the performance of EMIMFSI IL in PEO-LiTFSI system. They found the stable and uniform formation of Li dendrite between lithium and GPE during cycling (**Figure 12(a)**). They also evaluated the interfacial resistance of this passive layer and observed that except the initial few cycles almost stable interfacial resistance  $380 \Omega/\text{cm}^2$  was obtained throughout the cycling (**Figure 12(b)**).





**Figure 11.** Discharge capacity, efficiency (at C/10 rate) and capacity fading of the Li cell (a, b) Li/ (PEO + 20wt%LiTFSI + 20wt% trihexyltetradecylphosphonium TFSI)/LiNi<sub>0.6</sub>Mn<sub>0.2</sub>Co<sub>0.2</sub>O<sub>2</sub> and (c, d) Li/ (PEO + 20 wt% LiTFSI + 10 wt% EMIMFSI)/NCA.



**Figure 12.** (a) Voltage vs. time profile of lithium deposition and (b) evolution of interfacial resistances during cycling using GPE (PEO + 20wt% LiTFSI + 10wt% EMIMFSI).

Other reports on the electrochemical performance of Li batteries using PEO based GPEs are also tabulated in **Table 3**. All these analysis shows that the use of GPEs in Li battery maintains the cyclability and electrochemical stability of the Li battery much more compared to liquid solvents.

Therefore, from the above discussions it can be concluded that the IL based GPEs not only provide good ionic conductivity, flexibility and mechanical stability but also act as a potential candidate in order to enhance the capacity, cyclicity and safety to Li battery.

Polymer electrolytes	Li battery	C-rate	Capacity (mAh/g)	References
PEO <sub>20</sub> LiTFSI[Pyr13TFSI] <sub>1.27</sub>	Li/LiFePO <sub>4</sub> at RT	C/10	115 at 20 cycles	[54]
PEO <sub>20</sub> LiTFSI <sub>2</sub> [Pyr14TFSI] <sub>4</sub>	Li/NMC at 40°C	C/10	160 at 100th cycle	[55]
PEO <sub>20</sub> LiTFSI[Im12TFSI]	Li/LiFePO <sub>4</sub> at 50°C	C/5	110 at 20th cycle	[56]
PEO <sub>20</sub> LiTFSI[Pip <sub>1.101</sub> TFSI]	Li/LiFePO <sub>4</sub> at RT	C/20	120 at 35th cycle	[57]
PEO <sub>20</sub> LiTFSI[Pip <sub>1.101</sub> TFSI]	Li/Li <sub>4</sub> Ti <sub>5</sub> O <sub>12</sub> at RT	C/20	150 at 40th cycle	[57]
PEO <sub>20</sub> LiTFSI <sub>2</sub> [Pyr14TFSI] <sub>4</sub>	Li/LiFePO <sub>4</sub> at 40°C	C/10	140 at 450th cycle	[58]
PEO + 20wt%LiTFSI + 20wt%ThdpTFSI	Li/NMC622 at RT	C/10	148 at 150th cycle	[52]
PEO <sub>20</sub> LiTFSI <sub>2</sub> [Pyr14TFSI] <sub>2</sub>	Li/LiFePO <sub>4</sub> at 40°C	C/5	160 at 180th cycle	[59]
PEO-LiTFSI-10wt%EMIMFSI	Li/NCA at RT	C/10	175 at 200th cycle	[50]
PEO + LiFSI + 7.5 wt.% EMIMFSI	Li/LiFePO <sub>4</sub> at RT	C/20	143 at 100th cycle	[36]
PEO + LiTFSI + 12.5%EMIMTFSI	Li/LiMn <sub>2</sub> O <sub>4</sub> at RT	C/10	120 at 10th cycle	[42]
PEO + 20%LiTFSI + 30%(1-butyl 3-methyl pyridiniumTFSI)	Li/LiFePO <sub>4</sub> at 40°C	C/10	160 at 25th cycle	[51]
PEO + 20%LiFSI + 10%PYR13FSI	Li/GO-LiFePO <sub>4</sub> at rt	C/10	163 at 100th cycle	[49]
PEO + 20wt%LiTFSI + 20 wt% BMIMTFSI	Li/LiMn <sub>2</sub> O <sub>4</sub> at RT		140 μAh/cm <sup>2</sup> at 25 cycle	[48]

**Table 3.**  
*Electrochemical performance of Li batteries using IL based GPEs.*

7. Conclusion

In summary, many approaches have been proposed to enhance the conductivity of PEO based polymer electrolyte below melting temperature. Among them, ionic liquid based GPEs are considered as the most promising approach. These GPEs enhance the ionic conductivity, thermal and electrochemical stability of the polymer electrolytes. They provide better electrode-electrolyte contact, mechanical stability and safety to Li batteries. Because of having enough mechanical stability, they are able to suppress the undesirable dendrite formation and help to provide safer Li battery. Also, due to their high electrochemical stability, they can be used in high voltage and energy density batteries. In these batteries, they show good electrochemical and cyclic stability as well as offer flexibility and safety. Therefore, these IL based GPEs can be considered as a potential candidate for replacement of liquid electrolyte in Li batteries.

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Conflict of interest

The authors declare no conflict of interest.

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