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## Synthesis and characterization of SrBi<sub>4</sub>Ti<sub>4</sub>O<sub>15</sub> ferroelectric filler based composite polymer electrolytes for lithium ion batteries

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# Synthesis and characterization of SrBi<sub>4</sub>Ti<sub>4</sub>O<sub>15</sub> ferroelectric filler based composite polymer electrolytes for lithium ion batteries

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## Summary

Composite polymer electrolytes (CPEs) based on poly (ethylene oxide) (PEO) (Mol.Wt ~  $6 \times 10^5$ ) complexed with LiN(CF<sub>3</sub>SO<sub>2</sub>)<sub>2</sub> lithium salt and SrBi<sub>4</sub>Ti<sub>4</sub>O<sub>15</sub> ferroelectric ceramic filler have been prepared as films. Citrate gel technique and conventional solid state technique were employed for the synthesis of the ferroelectric fillers in order to study the effect of particle size of the filler on ionic conductivity of the polymer electrolyte. Characterization techniques such as X-ray diffraction (XRD), differential thermal analysis (DTA), scanning electron microscopy (SEM) and temperature dependant DC conductivity studies were taken for the prepared polymer composite electrolytes. The broadening of DTA endotherms on addition of ceramic fillers to the polymer salt complex indicated the reduction in crystallinity. An enhancement in conductivity was observed with the addition of SrBi<sub>4</sub>Ti<sub>4</sub>O<sub>15</sub> as filler to the (PEO)<sub>8</sub>-LiN(CF<sub>3</sub>SO<sub>2</sub>)<sub>2</sub> polymer salt complexes. Among the investigated samples (PEO)<sub>8</sub>-LiN(CF<sub>3</sub>SO<sub>2</sub>)<sub>2</sub> +10 wt% SrBi<sub>4</sub>Ti<sub>4</sub>O<sub>15</sub> (citrate gel) polymer composite exhibits a maximum conductivity.

## Introduction

Studies on polymer electrolytes have attracted global attention due to their potential applications in high energy density batteries, electrochromic devices and in sensors [1, 2]. The lithium polymer battery are highly desirable due to the characteristics such as inherent low safety risks and their ability to be formed into thin film structures of large surface area and yielding high energy density cells that are light weight and flexible [1]. Much of the efforts to date have focused on poly (ethylene oxide) (PEO) as the host material for solid polymer electrolyte. The major disadvantage with these polymer electrolytes is the insufficient ionic conductivity at room temperature and poor mechanical properties. Many modifications such as addition of inorganic fillers [3-14] and organic plasticizers have been carried out to improve the mechanical properties and ionic conductivity of the polymer electrolytes. Especially ferroelectric material BaTiO<sub>3</sub> in PEO has been reported to effectively increase the electrical

conductivity at lower temperatures and also decreases the interfacial resistance between the lithium anode and the polymer electrolyte [15]. In the present work, composite polymer electrolytes (CPEs) based on poly (ethylene oxide) (PEO) (Mol.Wt  $\sim 6 \times 10^5$ ) complexed with  $\text{LiN}(\text{CF}_3\text{SO}_2)_2$  lithium salt and  $\text{SrBi}_4\text{Ti}_4\text{O}_{15}$  ferroelectric ceramic filler have been synthesized. The ferroelectric fillers dispersed in the polymer electrolytes have been prepared by conventional solid-state reaction technique and citrate gel technique [16] to study the effect of particle size of the filler on ionic conductivity of the polymer electrolyte. The particle size of the filler is expected to have a wide influence on the ionic conductivity of the polymer electrolytes. These CPEs have been characterized using X-ray diffraction (XRD), differential thermal analysis (DTA), scanning electron microscopy (SEM) and temperature dependant DC conductivity techniques. For convenience the  $\text{SrBi}_4\text{Ti}_4\text{O}_{15}$  filler prepared through solid-state technique and citrate gel technique will hereafter be called as SBT (SST) and SBT (CIT), respectively.

## Experimental

### *Preparation of $\text{SrBi}_4\text{Ti}_4\text{O}_{15}$ (SBT) ferroelectric fillers*

The ferroelectric ceramic material  $\text{SrBi}_4\text{Ti}_4\text{O}_{15}$  was prepared by a conventional solid-state technique as described in our previous work [17]. The starting materials (AR grade) used for the preparation of SBT (CIT) were  $\text{SrCl}_2 \cdot 6\text{H}_2\text{O}$ ,  $\text{Bi}(\text{NO}_3)_3 \cdot 5\text{H}_2\text{O}$ , and  $\text{TiCl}_4$ . Citric acid was used as a chelating agent.  $\text{TiCl}_4$  was dissolved in ice-cold distilled water to form  $\text{TiOCl}_2$ . Weighted quantities of  $\text{SrCl}_2 \cdot 6\text{H}_2\text{O}$  in distilled water and  $\text{Bi}(\text{NO}_3)_3 \cdot 5\text{H}_2\text{O}$  in dil. HCl was added to the solution containing required amount of  $\text{TiCl}_4$  and citric acid. The solution was then placed in a water bath and maintained at  $60^\circ\text{C}$  and finally heated to  $100^\circ\text{C}$ . The pH of the solution was not maintained, as there was no precipitation during mixing. After the evaporation of water a slight yellowish gel was formed which was then transferred to a furnace and heated at  $250^\circ\text{C}$ . The gel started to swell producing a foamy precursor. This foam consisted of very light homogenous flakes of very small particle size. Intermediate grinding was done followed by further decomposing the precursor at  $800^\circ\text{C}$ .

### *Preparation of PEO- $\text{LiN}(\text{CF}_3\text{SO}_2)_2$ + $\text{SrBi}_4\text{Ti}_4\text{O}_{15}$ polymer composites*

The standard solution cast technique was used to prepare CPE films of thickness 200-250  $\mu\text{m}$  using acetonitrile (AR grade) as the common solvent. Poly (ethylene oxide) (Aldrich) of mol. wt. ( $\sim 6 \times 10^5$ ) and lithium bis (trifluoromethyl sulfonyl) imide [ $\text{LiN}(\text{CF}_3\text{SO}_2)_2$ ] (LiTFSI) salt (Aldrich) purchased were used without further purification. The lithium salt and SBT were dried in a furnace overnight at  $120^\circ\text{C}$  and  $200^\circ\text{C}$ , respectively, to remove the moisture prior to use.

Weighted quantity of PEO and  $\text{LiN}(\text{CF}_3\text{SO}_2)_2$  were first dissolved in acetonitrile and stirred thoroughly for 3 to 4 hrs at room temperature. The viscous solution was cast onto a teflon plate and allowed to dry at room temperature over night. The films were finally dried under vacuum for 3-4 days to evaporate any residual solvent. The O:Li ratio was varied in  $(\text{PEO})_x\text{-LiN}(\text{CF}_3\text{SO}_2)_2$  polymer salt complex for different values of x (where, x = 8, 16, 20 and 30). The filler dispersed composite polymer electrolytes were also prepared in the same manner as mentioned above with weighted quantity of

$\text{SrBi}_4\text{Ti}_4\text{O}_{15}$  added to the polymer salt complex. Thus, the compositions of the CPE films are represented as  $(\text{PEO})_8\text{-LiN}(\text{CF}_3\text{SO}_2)_2 + y$  SBT (SST and CIT) where ( $y = 0, 7.5, 10, 12.5$  and  $15$  wt%).

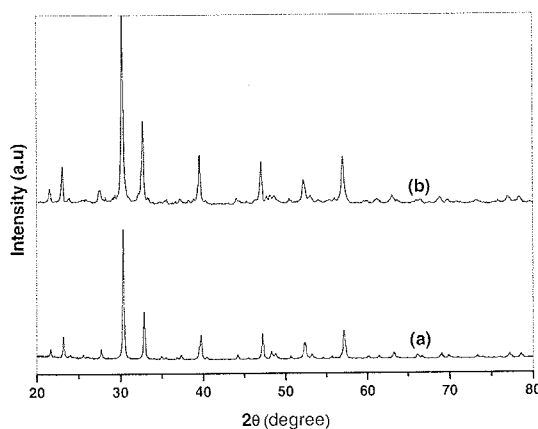
### Materials characterization

The XRD pattern for the ceramic fillers and CPEs were taken with a Philips 1730 diffractometer using  $\text{Cu K}\alpha$  radiation at a scan rate of  $1^\circ \text{ min}^{-1}$ . The DTA thermograms for PEO and  $(\text{PEO})_8\text{-LiN}(\text{CF}_3\text{SO}_2)_2 + y$  SBT (SST & CIT) ( $y=0, 7.5, 10, 12.5$  and  $15$  wt%) were recorded with a Linseis DTA instrument in the temperature range from  $30$  to  $120^\circ\text{C}$  at a heating rate of  $5^\circ\text{C}$  per min. The amount of each sample taken was  $\sim 10$  to  $12$  mg. The SEM images were taken for the ceramic fillers prepared by solid-state technique and citrate gel technique with a JEOL JSM-6460A to investigate the microstructure of the fillers. Temperature dependant DC conductivity studies were taken by casting the polymer samples on teflon moulds. Preparing the sample in a teflon mould ensure the intactness of the sample during conductivity measurement. The teflon mould was then placed in a specially designed vacuum chamber. Silver electrodes were used for the DC conductivity measurements carried out in vacuum for various temperature ranges starting from room temperature ( $27^\circ\text{C}$ ) to  $120^\circ\text{C}$ .

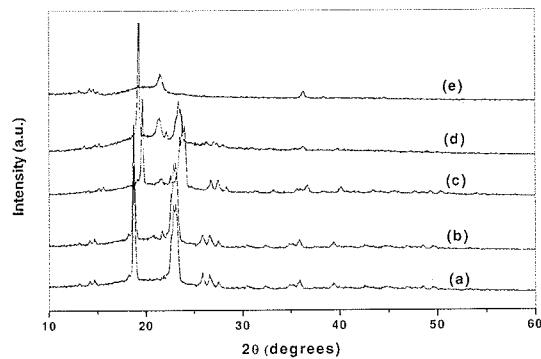
### Results and Discussion

#### XRD analysis

SBT belongs to the Aurivillius family of oxides and exhibit ferroelectric to paraelectric transition at elevated temperature. The XRD patterns of the ceramic fillers both prepared by solid state and citrate gel method are as shown in Fig. 1.

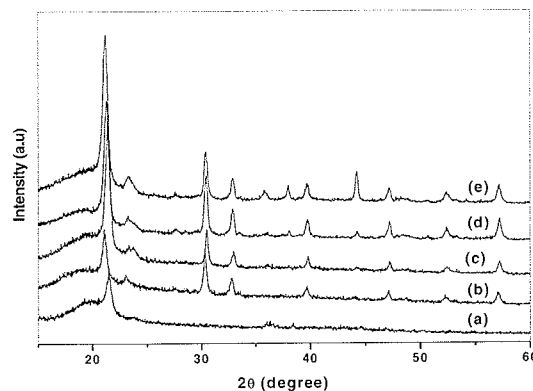


**Fig. 1.** XRD patterns of the  $\text{SrBi}_4\text{Ti}_4\text{O}_{15}$  prepared by (a) solid state technique and (b) citrate gel technique



**Fig. 2.** XRD patterns of (a) PEO and  $(\text{PEO})_x\text{-LiN}(\text{CF}_3\text{SO}_2)_2$  salt complex where (b)  $x = 30$ , (c)  $x = 20$ , (d)  $x = 16$  and (e)  $x = 8$

There is a slight increase in width of diffraction peaks of SBT (CIT) when compared to the diffraction peaks of SBT (SST), which reveals the reduction in particle size of the filler prepared by citrate gel technique when compared to SBT (SST). The position and intensities of dominant diffraction peaks of  $\text{SrBi}_4\text{Ti}_4\text{O}_{15}$  observed in Fig. 1 matches well with the JCPDS card number 43-0973. Fig. 2 displays the X-ray diffraction patterns of  $\text{PEO-LiN}(\text{CF}_3\text{SO}_2)_2$  complexed films. The characteristic peaks for crystalline PEO are found between  $15^\circ$  and  $30^\circ$ . As the salt concentration increases the sharp diffraction peaks corresponding to PEO becomes less prominent accompanying the decrease in intensity and gradual broadening of the diffraction peaks. At an O/Li ratio of 8:1 the characteristic peak of PEO diminishes and the peak at  $21^\circ$  corresponding to the polymer salt complex begins to gain intensity. This observation is in accordance with previous studies on PEO with changing salt concentration [18]. The XRD patterns of the polymer salt complex with different wt % of SBT (citrate gel) are shown in Fig. 3.

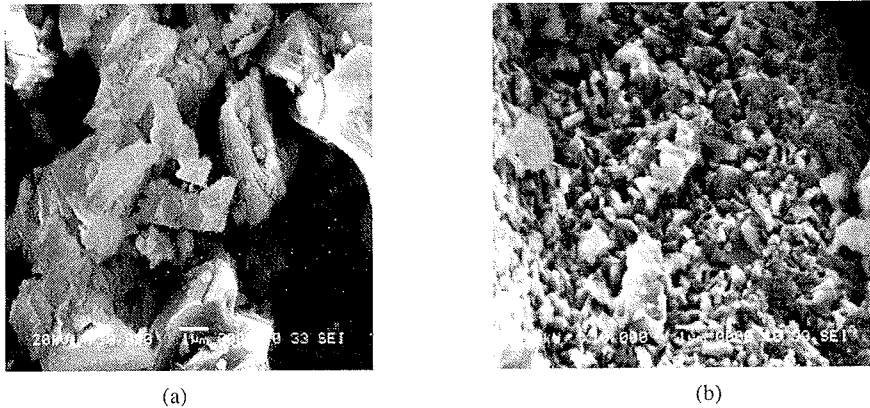


**Fig. 3.** XRD patterns of  $(\text{PEO})_8\text{-LiN}(\text{CF}_3\text{SO}_2)_2 + y \text{ SBT (CIT)}$  salt complex where (a)  $y = 0$  (b)  $y = 7.5$ , (c)  $y = 10$ , (d)  $y = 12.5$  and (e)  $y = 15 \text{ wt}\%$

As the wt% of the filler increases the peaks corresponding to the SBT becomes prominent in the XRD pattern. The general broad nature of the prominent diffraction peaks shows the reduction in the degree of crystallinity of the composite polymer electrolytes.

#### *SEM analysis*

The SEM images of SBT (SST) and SBT (CIT) are shown in Fig. 4. The sample prepared by conventional solid-state technique has a larger particle size of the order of  $3\mu\text{m}$ . The SBT prepared by citrate gel method has much lesser particle size of approximately  $0.5\mu\text{m}$  and moreover the particle sizes are homogeneously distributed. The reduction of particle size is expected to have considerable changes in the conductivity and the crystalline nature of the polymer salt complex.

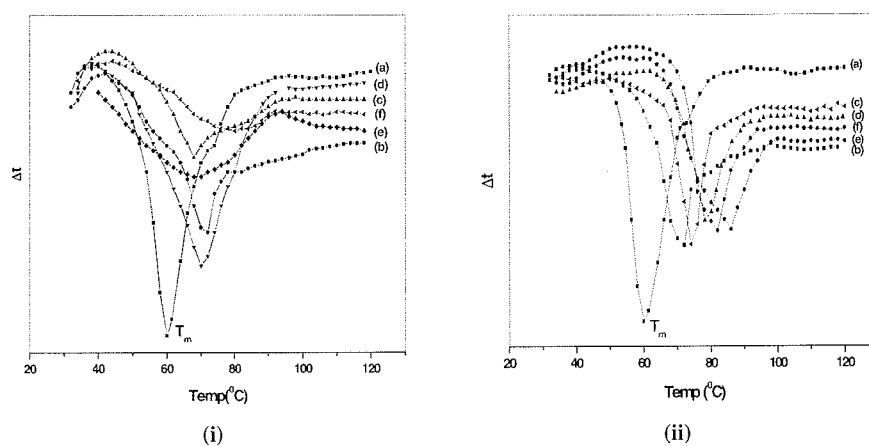


**Fig. 4.** SEM micrographs of  $\text{SrBi}_4\text{Ti}_4\text{O}_{15}$  filler synthesized by a) solid state technique and (b) citrate gel technique

#### *Differential thermal analysis*

The differential thermal analysis thermograms of PEO and  $(\text{PEO})_8\text{-LiN}(\text{CF}_3\text{SO}_2)_2 + y$  SBT (SST) ( $y=0, 7.5, 10, 12.5$  and  $15$  wt%) shown in Fig. 5(i), revealed the changes in the melting temperature ( $T_m$ ) of the polymer salt complex as a result of the addition of ferroelectric filler in the polymer matrix. The predominant endothermic peak ( $T_m$ ) is attributed to the melting of the crystalline phase of PEO. The melting temperature increases from  $60$  to  $70^\circ\text{C}$  for the pure PEO to  $(\text{PEO})_8\text{-LiN}(\text{CF}_3\text{SO}_2)_2$  complex. Addition of the filler with wt% of  $7.5, 10$  and  $12.5$  to  $(\text{PEO})_8\text{-LiN}(\text{CF}_3\text{SO}_2)_2$  polymer salt complex does not show any significant increase in  $T_m$  however there is noticeable changes in  $T_m$  for  $15$  wt% of the filler.

As the filler wt% increases there is a significant broadening of the endotherms and is maximum for  $15$  wt% of the filler. The DTA thermograms of PEO and  $(\text{PEO})_8\text{-LiN}(\text{CF}_3\text{SO}_2)_2 + y$  SBT (CIT) ( $y=0, 7.5, 10, 12.5$  and  $15$  wt%) are shown in Fig. 5(ii).



**Fig. 5.** DTA thermograms of (a) PEO and (PEO)<sub>8</sub>: LiN(CF<sub>3</sub>SO<sub>2</sub>)<sub>2</sub> + y SBT (SST) salt complex where (b) y = 0 (c) y = 7.5, (d) y = 10, (e) y = 12.5 and (f) y = 15 for (i) SBT (SST) and (ii) SBT (CIT)

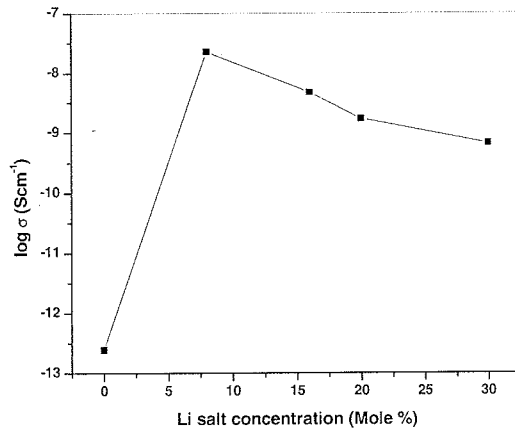
There is a notable increase in the melting temperature  $T_m$  of the CPEs as the filler weight percent is increased except for 15wt% for which a slight decrease in  $T_m$  is observed compared with  $y = 12.5$  wt%. The broad nature of the endotherms especially for (PEO)<sub>8</sub>-LiN(CF<sub>3</sub>SO<sub>2</sub>)<sub>2</sub> +15 wt% of SBT (SST) and (PEO)<sub>8</sub>-LiN(CF<sub>3</sub>SO<sub>2</sub>)<sub>2</sub> +12.5 wt% of SBT (CIT) is an indication for an increase in the amorphous regions of the CPEs.

The observed DTA behavior proves the characteristic role of the filler in inhibiting crystallization, which thereby reduces the volume fraction of the crystallization in the polymer salt complex. The broadening of the endotherm also suggests that there is a decrease in the flexibility of the polymer. This might be due to the increase in the intramolecular attraction, as they form transient cross-links in the polymer matrix as a result of which the mechanical stability may increase.

#### *Conductivity measurements*

Fig. 6 shows the room temperature DC ionic conductivity plot for PEO and (PEO)<sub>x</sub>-LiN(CF<sub>3</sub>SO<sub>2</sub>)<sub>2</sub> (O/Li=8,16,20 and 30). As the mole ratio of lithium salt is increased there is a significant increase in the room temperature conductivity of the electrolytes and reaches a maximum of  $2.29 \times 10^{-8} \text{ Scm}^{-1}$  for a [O]: [Li] ratio of 8:1.

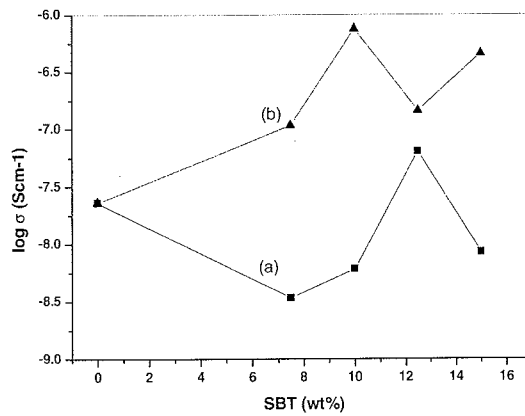
The increase in conductivity for the mole ratio 8:1 is related to the high dissociation of the salts and increasing concentration of the charge carriers. This observation is consistent with earlier reports on polymer electrolytes derived from PEO and Li salts [19]. When the ferroelectric fillers SBT (SST) and SBT (CIT) were added to the polymer salt complex we observed an enhancement in conductivity for the composite electrolytes. Przulski et al [20] suggested that a highly conductive layer is formed in the neighbourhood of the filler particles through the Lewis-acid base reaction. In such composite polymer electrolytes, the permanent dipole of the ferroelectric SrBi<sub>4</sub>Ti<sub>4</sub>O<sub>15</sub>



**Fig. 6.** Ionic conductivity Vs  $\text{LiN}(\text{CF}_3\text{SO}_2)_2$  salt concentration for PEO and  $(\text{PEO})_x$ ;  $\text{LiN}(\text{CF}_3\text{SO}_2)_2$  ( $x = 8, 16, 20, 30$ ) polymer salt complex measured at room temperature

could act as an origin of the Lewis acid base interaction and form a highly conductive layer by strong interaction with the other constituents in the polymer electrolytes, showing higher ionic conductivity than the pristine polymer electrolyte [21,22].

Comparison of the room temperature DC conductivity for different wt% of the SBT (both prepared by SST and CIT method) in  $(\text{PEO})_x$ - $\text{LiN}(\text{CF}_3\text{SO}_2)_2$  is shown in Fig. 7. It can be clearly seen that the SBT prepared by citrate gel ( $0.5\mu\text{m}$ ) method exhibits a better conductivity than the SBT prepares by a solid state method ( $3\mu\text{m}$ ) due to the reduction in particle size of the filler. Initially for a low wt% of the SBT (SST) there is a decrease in the conductivity and then as the filler wt% is increased the conductivity gradually increases and reaches a maximum of  $0.64 \times 10^{-7} \text{ Scm}^{-1}$  for 12.5 wt% of the filler.



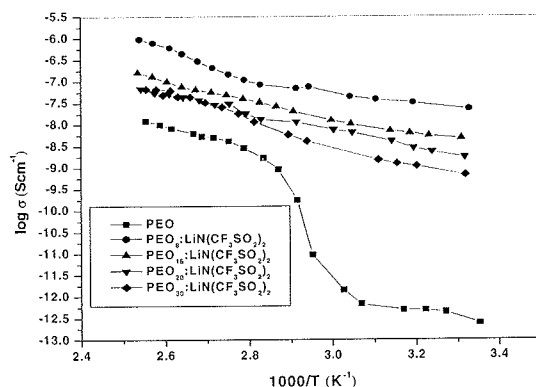
**Fig. 7.** Comparison of room temperature ionic conductivity of the CPEs for different wt% of the filler prepared by (a) solid state technique and (b) citrate gel technique



The initial decrease in the conductivity for SBT (SST) filler might be due to inhomogeneous distribution of the ceramic filler in the polymer matrix and thus reducing the conductivity. The ionic conductivity for SBT (CIT) polymer films steadily increase with increase in wt% of the filler and reaches a maximum of  $0.75 \times 10^{-6} \text{ Scm}^{-1}$  for 10 wt% of the filler. We observed that the conductivity value ( $\sim 10^{-6} \text{ Scm}^{-1}$ ) observed for composite polymer electrolytes with  $\text{SrBi}_4\text{Ti}_4\text{O}_{15}$  ferroelectric fillers prepared by citrate gel technique was higher than the conductivity observed by Park et. al. [23] for composite polymer electrolyte with  $\text{BaTiO}_3$  filler ( $\sim 10^{-7} \text{ Scm}^{-1}$ ) at ambient temperature.

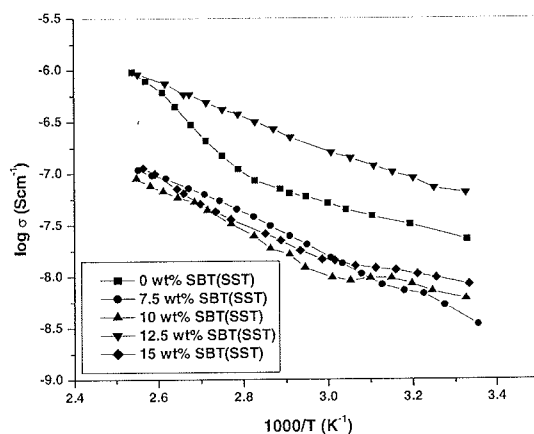
The addition of the filler with reduced particle size tends to uniformly contribute to the reduction in crystallinity of the polymer matrix by promoting effectively the dissociation of ion-ion pairs and ion clusters in the polymer electrolytes with the help of the strong electrostatic interaction caused by permanent dipoles. The decrease in ionic conductivity for higher wt% of the fillers might be due to the fact that inorganic fillers tend to impede ionic movement by acting as an insulator and an agglomerator.

The temperature dependent DC ionic conductivity for the prepared composite polymer films was taken from room temperature ( $27^\circ\text{C}$ ) to  $120^\circ\text{C}$ . Arrhenius plots were plotted for the PEO,  $(\text{PEO})_x\text{-LiN}(\text{CF}_3\text{SO}_2)_2$  ( $x = 8, 16, 20$  and  $30$ ) and  $(\text{PEO})_8\text{-LiN}(\text{CF}_3\text{SO}_2)_2 + y\text{SBT}$  (SST& CIT) ( $y = 0, 7.5, 10, 12.5$  and  $15$  wt%). As the temperature is increased from room temperature to  $120^\circ\text{C}$  as shown in Fig. 8, there is a steep rise at around  $60^\circ\text{C}$  for pure PEO film, which is due to the melting of the crystalline regions in the PEO matrix.



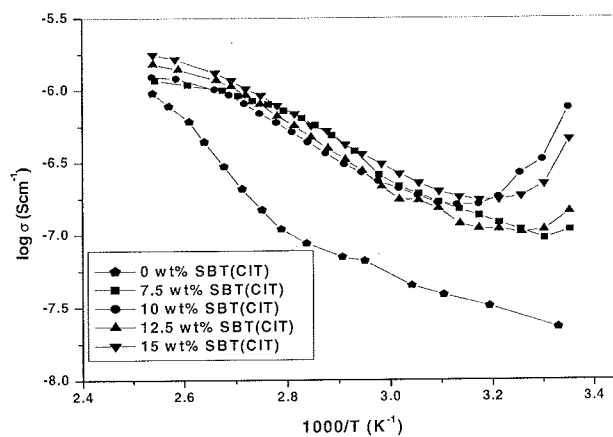
**Fig. 8.** Temperature dependent DC conductivity plots for pure PEO and  $(\text{PEO})_x\text{-LiN}(\text{CF}_3\text{SO}_2)_2$  for different values of  $x$  ( $x = 8, 16, 20$  and  $30$ )

As the salt concentration increases the non-linear region shifts to a higher temperature with an increase in conductivity for the entire temperature range. The Arrhenius plot shown in Fig. 9 indicated that except for  $(\text{PEO})_8\text{-LiN}(\text{CF}_3\text{SO}_2)_2 + 12.5$  wt% of SBT(SST) the temperature dependant conductivity for other wt% of SBT (SST) are much lower than the  $(\text{PEO})_8\text{-LiN}(\text{CF}_3\text{SO}_2)_2$  films. At  $\sim 120^\circ\text{C}$  both the  $(\text{PEO})_8\text{-LiN}(\text{CF}_3\text{SO}_2)_2$  and  $(\text{PEO})_8\text{-LiN}(\text{CF}_3\text{SO}_2)_2 + 12.5$  wt% of SBT(SST) exhibited a similar conductivity.



**Fig. 9.** Temperature dependant DC conductivity plots for  $(\text{PEO})_8: \text{LiN}(\text{CF}_3\text{SO}_2)_2 + y$  SBT (SST) CPEs for ( $y = 0, 7.5, 10, 12.5, 15$ ) wt% of the filler

The high temperature conductivity profiles for  $(\text{PEO})_8\text{-LiN}(\text{CF}_3\text{SO}_2)_2 + y$  SBT (CIT) ( $y = 7.5, 10, 12.5$  and  $15$  wt% did not exhibit much significant changes at  $120^\circ\text{C}$  as shown in Fig. 10.



**Fig. 10.** Temperature dependant DC conductivity plots for  $(\text{PEO})_8: \text{LiN}(\text{CF}_3\text{SO}_2)_2 + y$  SBT (CIT) CPEs for ( $y = 0, 7.5, 10, 12.5, 15$ ) wt% of the filler

Comparatively the  $(\text{PEO})_8\text{-LiN}(\text{CF}_3\text{SO}_2)_2 + 15\text{wt}\%$  of SBT (CIT) exhibit a higher conductivity at  $120^\circ\text{C}$ . The temperature dependant conductivity profile for  $(\text{PEO})_8\text{-LiN}(\text{CF}_3\text{SO}_2)_2 + 10\text{wt}\%$  of SBT(CIT) and  $(\text{PEO})_8\text{-LiN}(\text{CF}_3\text{SO}_2)_2 + 12.5\text{wt}\%$  of SBT(CIT) exhibited a non linear region upto  $40^\circ\text{C}$  with a drop in conductivity above which the conductivity steadily increased.

## Conclusion

A lithium ion conducting polymer electrolyte system  $(\text{PEO})_8\text{-LiN}(\text{CF}_3\text{SO}_2)_2 + y \text{SrBi}_4\text{Ti}_4\text{O}_{15}$  (SST and CIT, where  $y = 0, 7.5, 10, 12.5$  and  $15 \text{ wt\%}$ ) has been prepared by solvent casting technique in film form and characterized by XRD and DTA. SEM images reveal that the  $\text{SrBi}_4\text{Ti}_4\text{O}_{15}$  prepared by citrate gel method has much lesser particle size than the one prepared by conventional solid-state reaction. The room temperature DC conductivity shows that SBT prepared by citrate gel method exhibits a better conductivity due to the reduction in particle size of the filler. The ionic conductivity for SBT (SST) and SBT (CIT) dispersed polymer composites reaches a maximum of  $0.64 \times 10^{-7} \text{ Scm}^{-1}$  (12.5 wt%) and  $0.75 \times 10^{-6} \text{ Scm}^{-1}$  (10 wt%) at room temperature respectively. Temperature dependant DC conductivity for SBT (SST) polymer films showed a decrease in conductivity over the entire temperature range when compared to  $(\text{PEO})_8\text{-LiN}(\text{CF}_3\text{SO}_2)_2$  films except for  $(\text{PEO})_8\text{-LiN}(\text{CF}_3\text{SO}_2)_2 + 12.5 \text{ wt\%}$  of SBT (SST) which showed an increase in conductivity. The temperature dependant conductivity profiles for SBT (CIT) polymer films showed an increase in conductivity with the addition of the filler and exhibit a maximum conductivity of  $0.176 \times 10^{-5} \text{ Scm}^{-1}$  at  $120^\circ\text{C}$  for 15 wt % of the filler.

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