



Impacts of ceramic filler and the crystallite size of polymer matrix on the ionic transport properties of lithium triflate/poly (vinylidene fluoride-co-hexafluoropropene) based polymer electrolytes



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ABSTRACT

Solid polymer electrolytes (SPEs) have wide-scale applications in the fields of energy storage (capacitors and batteries), power conversion (fuel cells), electrochemical sensing etc. In the present work, poly (vinylidene fluoride-co-hexafluoropropene) (PVDF-HFP) based SPEs with varying concentration of lithium triflate and composite solid polymer electrolytes (CSPEs) composed of BaTiO₃ in addition to the optimised loading of lithium triflate in PVDF-HFP matrix are synthesized by solution casting technique. SPE with 10 wt.% lithium triflate shows highest DC conductivity of $2.19 \times 10^{-6} \text{ S cm}^{-1}$ at 300 K, which enhances to $8.89 \times 10^{-6} \text{ S cm}^{-1}$ by adding 4 wt.% of BaTiO₃. X-Ray diffraction (XRD) and scanning electron microscopy (SEM) analysis shows that the reduction in crystallite size of the polymeric matrix assists in enhancing the conductivity. Hence, the crystallite size, by altering the arrangement of the non-conducting polymeric chains, affects the ionic conductivity of the composites significantly.

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1. Introduction

The electrochemical power conversion devices such as primary batteries, fuel cells, electrochemical solar cells, etc., which convert chemical/solar energy to electric energy, and the electrochemical power storage devices such as secondary batteries, supercapacitors, etc., which store electric energy through electrochemical routes, are the essential candidates fulfilling our daily energy requirements [1–4]. These electrochemical power storage/conversion devices consist of two electrodes separated by an electrolyte that facilitates the charge transport between the electrodes through conduction of ions. High ionic conductivity, chemical inertness and stability under the device environment, etc. are the material properties essential for proper functioning of the electrolytes. Based on the physical state, electrolytes can be classified into two broad categories, namely the liquid electrolytes and the solid electrolytes. Although the liquid electrolytes have ionic conductivities higher than those of the solid electrolytes,

disadvantages such as corrosion of the electrodes, limited temperature range of operation, formation of dendrites, leakage, unsuited to miniaturization, etc. make them inappropriate for various advanced applications [5]. Therefore, solid electrolytes that overcome these disadvantages are being used in advanced applications such as batteries, fuel cells, gas sensors, ion selective membranes etc. [6]. Various types of materials including ceramics, glassy solids, polymers, etc. and their composites are being used as solid electrolytes in different applications [7–11]. Among these, solid polymer electrolytes (SPEs) are one of the most promising options owing to their lower manufacturing cost, adaptability to a wide variety of packaging shapes, reliability, ruggedness and better flame-retardant properties [6,12–17]. Since their discovery, SPEs have undergone various stages of development from dry SPEs to polymer gel electrolytes, till recently to composite SPEs (CSPEs).

Lithium based polymer electrolytes consist of polymer matrix with lithium salt filler. The beginning of lithium polymer electrolyte goes back to 1973 with the work by Fenton et al. suggesting polyethylene oxide (PEO) lithium system as the ionic conducting polymer electrolyte [18]. PEO has a high electrochemical stability in comparison with other polyethers and possesses high degree of crystallinity, due to which the ionic conductivity of

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PEO-based electrolytes varies from $10^{-4} \text{ S cm}^{-1}$ to $10^{-8} \text{ S cm}^{-1}$ with temperature change from 40 to 100°C . By 1980, it was well known that the amorphous phase is responsible for ionic conduction. As the ionic conductivity of SPEs at room temperature is very low compared to the liquid electrolytes, significant research efforts have been focused on improving the ionic conductivity of SPEs. The most important strategy employed to improve the ionic conductivity of SPEs is to increase their amorphous phase by using plasticizers or ceramic fillers [19]. To be applicable for practical applications, SPEs should have a ionic conductivity $>10^{-4} \text{ S cm}^{-1}$, along with high chemical, electrochemical and thermal stabilities, high mechanical strength and an ionic transference number of ~ 1 [19].

A large variety of SPEs have been developed with different combinations of lithium salts and polymer host matrices i.e., conventional polymer-salt complex [20], plasticized polymer-salt complex [21], polymer gel electrolyte [22] and rubbery electrolyte

[23]. All these classes have some problems/disadvantages, for example, in conventional polymer-salt complex, conductivity and Li^+ transference number decreases with increase in lithium ion salt [7,24–27]. Similarly, the polymer gel electrolyte show poor mechanical strength, while in the rubbery electrolytes, dissolved salts have a tendency to crystallize [28]. To CSPEs, on the other hand, being composed of SPEs filled with inert or ionic conductive fillers, have significant potential towards eliminating these issues.

Polyvinylidene fluoride-hexafluoropropylene (PVDF-HFP) is a prospective electrolyte material in lithium-ion batteries because of its attractive and interesting properties such as high dielectric constant of 8.4 [29,30]. Recently, the effect of various fillers (i.e. SiO_2 , Al_2O_3 and BaTiO_3) on the PVDF-HFP based composite polymer electrolytes based on lithium hexafluorophosphate (LiPF_6) has been studied. The study by Xiao et al. has revealed that the BaTiO_3 based composite polymer electrolyte shows highest conductivity because of its good interaction with the PVDF-HFP [31]. It is known

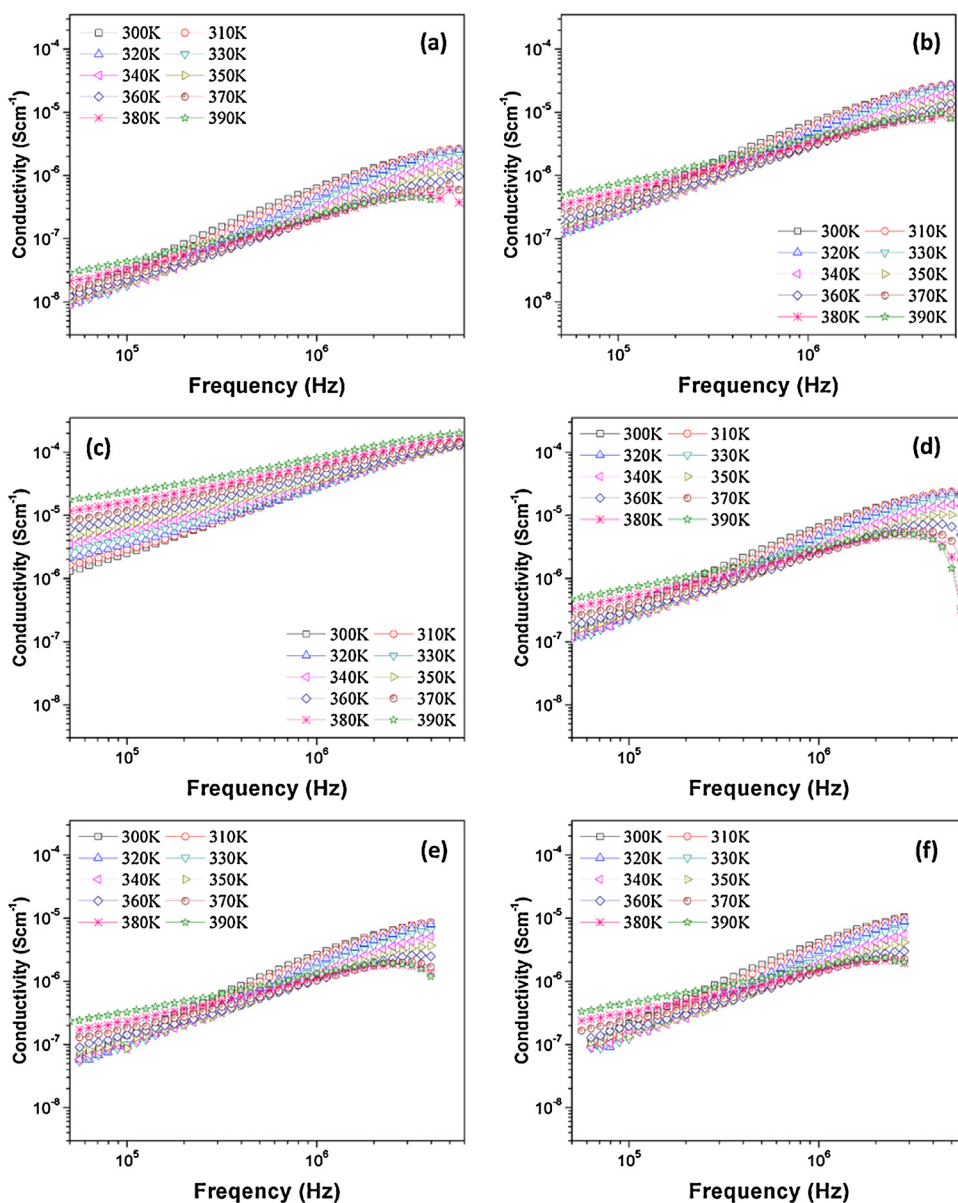


Fig. 1. AC conductivity spectra over a range of temperatures (K) of (a) pure polymer and lithium triflate salt/PVDF-HFP SPEs with (b) 5, (c) 10, (d) 15, (e) 20 and (d) 25 wt.% of lithium triflate.

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