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#### ABSTRACT

This review article provides a deep insight into the ion conduction mechanism in polymer electrolytes (PEs). The concepts of different categories of polymer electrolytes are discussed. The significance of the existence of functional (polar) groups on the backbone of host polymers, which are used in polymer electrolytes, is well explained. The working principle of electrical impedance spectroscopy (EIS) is overviewed. The relationship between impedance plots and equivalent circuits, which are crucial for electrical characterization, is extensively interpreted. Based on the patterns of dc conductivity ( $\sigma_{dc}$ ) versus 1000/T, the ion transport models of Arrhenius and Vogel–Tammann–Fulcher (VTF) are discussed. Effects of coupling and decoupling between ionic motion and polymer segmental relaxation are analyzed. The important role of dielectric constant on cationic transport in PEs is also explained. The relationships existing between electrical and dielectric parameters are elucidated, which help interpret and understand the ion conduction mechanism. From the reported empirical curves of dc conductivity

vs. dielectric constant, the reformulated Arrhenius  $\left[\sigma_{dc}(T) = \sigma_0 \exp\left(-\frac{E_a}{k_B T_{ef}}\right)\right]$  equation is proposed. Finally, other important phenomena, occurring in polymer electrolytes, are shown to be understandable from the dielectric constant studies.

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## 1. Introduction

Various sources of alternative energy are continuously evolving to reduce the long-term dependence on oil, nuclear and other fossil fuels. The other environmentally friendly fuel cells, such as batteries, super capacitors and dye sensitized solar cells, are strong candidates for this reason [1]. The conception of polymer electrolytes (PEs) is a highly specialized and multidisciplinary field that covers the disciplines of electrochemistry, polymer science, organic and inorganic chemistry [2]. Dry solid polymer electrolytes (SPEs) have attracted great attention as safer alternatives to liquid electrolytes [3]. In the field of SPEs, a pioneering work was carried out by Wright et al. and cited by Singh and Bhat [4]. In their work, the dc conductivity of order of  $10^{-5}$  S/cm at

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330 K in highly crystalline polyethylene oxide-sodium thiocyanate (PEO–NaSCN) complexes was reported [4,5]. The SPEs are formed by inorganic salts dissolution in a polar polymer matrix. The choice of PEs in modern applications, such as high energy density batteries, electrochromic devices, sensors and fuel cells, was justified by studying their structural, morphological and electrical properties [6,7]. On the other hand, the choice of polymer hosts for PEs largely depends on two factors: first, the existence of polar (functional) groups with a large power of sufficient electron donor to form coordination with cations and, second, a low hindrance to bond rotation [2]. Fig. 1 shows the chemical structures of some important polymers that are widely used as host polymers in PEs.

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The good mechanical strength, ease of thin film fabrication with desirable shapes and the ability of forming good electrode/electrolyte contact are the main advantages of dry SPEs [6,8,9]. From the economical and commercial viewpoints, a low-cost membrane with good ionic conductivity, enhanced dimensional and mechanical stabilities are recent challenges to be invented. The main drawbacks of SPEs are their high crystallinity and low ionic

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**Fig. 1.** Chemical structures of some polar polymers widely used for polymer electrolytes: (a) Poly (ethylene oxide) (PEO), (b) Poly(vinyl alcohol) (PVA), (c) Poly(methyl methacrylate) (PMMA), (d) Poly(ε-caprolactone) (PCL), (e) Chitosan (CS), (f) Poly(vinylpyrrolidone) (PVP), (g) Poly(vinyl chloride) (PVC), and (h) Poly(vinylidene fluoride) (PVDF).

conductivity [10]. Polymer electrolytes comprise both crystalline and amorphous regions. It has been well reported that the ion transport occurs mainly in the amorphous region rather than the crystalline region, but the polymers host materials that used in PEs are often semi-crystalline [4,11]. Thus, to overcome the disadvantages and improve SPEs' conductivity, plasticizer, as one mostly applied method, has to be added to improve the ambient ionic conductivity. Through using plasticizers, the amorphous region and ion aggregates in PEs can be increased and dissociated, respectively, causing the dc electrical conductivity of SPE to be improved [12]. It has been established that the ionic conductivity in plasticized polymer electrolytes can be increased at the expense of decreased mechanical strength and vice versa [4,13]. In addition to high conductivity and a broad electrochemical stability window, PEs must exhibit good thermal and mechanical properties. These performances can be achieved by dispersing nanosized fillers into polymer electrolyte. Following the creative work of Weston and Steele [4,14], who have improved the ionic conductivity and mechanical stability of polymer electrolytes by adding Al<sub>2</sub>O<sub>3</sub> particles, nanocomposite SPEs have been broadly studied. A complete understanding of the effects of inorganic fillers on the ion transport, thermal, mechanical and electrochemical properties of PEs is still not reached [15]. From the above survey, it is clear that the dc electrical conductivity can be improved by incorporating the plasticizer or inorganic fillers into the SPEs. But yet, the ion conduction mechanism in solid plasticized and composite polymer electrolytes is not fully understood [4]. The main goal of this review article is to shed light on different types of PEs and ion transport models. Additionally, the necessary of reformulation of Arrhenius equation based on recent experimental achievements in this field is elucidated.

## 2. Classifications of polymer electrolyte

Polymer electrolytes have been proved to be promising materials in the research and development of electrochemical devices. Most of the research activities are devoted in the field of solid state electrochemistry, in which high ion-conducting materials are considered to be developed for the energy conversion and storage applications [16]. In this sense, PEs are a class of materials, which have been witnessed in the last 20 years by massive research efforts, to achieve systems with a good conductivity and an electrochemical stability [16,17]. On the basis of materials, the polymer electrolytes have been categorized into dry solid polymer electrolyte, plasticized polymer electrolytes, gel polymer electrolytes, and composite polymer electrolytes [18]. Download English Version:

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