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## Zirconium dioxide nanofilled poly(vinylidene fluoride-hexafluoropropylene) complexed with lithium trifluoromethanesulfonate as composite polymer electrolyte for electrochromic devices

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#### ABSTRACT

Poly(vinylidene fluoride-hexafluoropropylene) (PVdF-HFP) polymer electrolyte containing zirconium dioxide nanocrystals (ZrO<sub>2</sub>-NC) and lithium trifluoromethanesulfonate (LiCF<sub>3</sub>SO<sub>3</sub>) has been synthesized using the conventional solution casting method. The addition of ZrO<sub>2</sub>-NC into the polymeric substrate gave remarkable properties in terms of the electrolyte's ionic conductivity as well as its bulk mechanical strength. The enhanced amorphicity of the polymeric substrate due to ZrO<sub>2</sub> and the nanofiller's high dielectric constant make an excellent combination to increase the ionic conductivity (above  $10^{-4} \text{ S cm}^{-1}$ ). Increasing the nanofiller content raises the ionic conductivity of the electrolyte by two orders of magnitude of which the optimum is  $2.65 \times 10^{-4} \text{ S cm}^{-1}$  at  $13.04 \text{ wt% ZrO<sub>2</sub>-NC loading. Also, the Young's modulus, an indicator of electrolyte's spectroscopy, the electrolytes with <math>13.04\% \text{ ZrO<sub>2</sub>-NC}$  scanned from 200–800 nm wavelengths exhibited a maximum optical transmittance of 52.6% at  $10 \,\mu\text{m}$  film thickness. The enhanced conductivity, high mechanical strength and reasonable optical transmittance shown by our composite polymer electrolyte make an excellent electrolyte for future energy saving smart windows such as electrochromic devices.

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#### 1. Introduction

Electrochromic smart windows are characterized by their ability to control the amount of light transmission by the application of low electrical voltage thereby providing energy efficiency and indoor comfort in buildings. Electrochromic devices (ECDs) are generally composed of an electrolyte, electrochromic layer, counter electrode and electrical conductors. Electrolytes and electrochromic materials are the most vital and active components of an electrochromic device. Electrochromic materials have the ability to reversibly alter their light absorption properties within a specific range of spectral wavelength through redox reactions stimulated by low direct current (DC) potentials which result to a color change. This change is due to the generation of new absorption band in the visible region, infrared or in the microwave integrated with ion conducting electrolytes and some electrical contacts. The polymer electrolytes play a very essential role as the primary medium for ionic conduction in the ECD and it can be in the form of a liquid, gel or solid [2]. Two of the major functions of electrolyte in electrochemical systems are to promote mutual ionic flow and exchange between the electroder while keeping them from direct electrical contact

region [1]. For this to take place, the electrochromic layer must be

systems are to promote mutual ionic flow and exchange between the electrodes while keeping them from direct electrical contact and to avoid electronic charge flow [2,3]. Most electrolytes are composed of solvents and salts which ionize to provide ions in the system [4]. Electrolytes have different classes and each has its own strengths and drawbacks [5,6]. They are generally classified into liquid electrolytes, ceramic electrolytes, solid inorganic electrolytes and polymer electrolytes [7–11]. Based on previous studies, polymer electrolytes have been preferred and were tailored for advanced electrochemical systems especially for lithium-ion batteries and electrochromic applications [12,13]. They are preferred because of their processibility, mechanical strength, stability, flexibility, and wide range of working temperature [1].







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Table 1	
Weight ratio of components for the synthesis of composite polymer electroly	/tes.

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Polymer electrolytes are very vital in ECDs for they dictate the overall performance of the device [14–16]. Thus, a well-tuned formulation is very essential for high efficiency and for longer cycle life of the device [17–21]. For a polymer electrolyte to be suitable for electrochromic applications, it has to be highly ion conductive, has a good electron donating ability, has low bond rotation energy, thermally and electrochemically stable, has high optical transparency, able to cover a wide range of potential, able to be confined into specific areas, and it has to be cheap but environmentally friendly [14–24].

Polymer electrolytes can further be categorized into solids, gels, polyelectrolytes, and composites [1]. Solid polymer electrolytes are

preferred due to chemical and electrochemical stability, flexibility and mechanical versatility, and ease of processing they offer. However, these types generally give a low ionic conductivity. Gel polymer electrolytes, on the other hand, have higher ambient ionic conductivity but are mechanically weak. Polyelectrolytes, usually, are not sufficiently flexible and they exhibit ionic conductivities around or below 10<sup>-6</sup> S cm<sup>-1</sup> [1]. Composite polymer electrolytes exhibit highly improved ionic conductivity compared to the former three types. The addition of inorganic fillers like zeolites. Al<sub>2</sub>O<sub>3</sub>, SiO<sub>2</sub> and MgO has dramatically increased the ionic conductivity of electrolytes. Furthermore, the interfacial stability and the mechanical strength of the composites were also enhanced. The polymer matrices that were investigated in the recent years are polyethylene oxide (PEO), polyacrylonitrile (PAN), polymethyl methacrylate (PMMA), and polyvinylidene fluoride (PVdF) [7,25–29]. PVdF has gained attention and been used recently as electrolyte for electrochromic systems due to its many advantages like high mechanical strength and its high dielectric constant which aids in the ionization of the salt giving a higher charge density. However, PVdF does not work well with lithium ions because of its semi-crystalline property [1]. The mobility of the ions is suppressed in this type of matrix which consequently limits



Fig. 1. X-ray diffraction patterns of (a) PVdF-HFP and (b) ZrO<sub>2</sub>.

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