



# A novel plastic crystal composite polymer electrolyte with excellent mechanical bendability and electrochemical performance for flexible lithium-ion batteries



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## ABSTRACT

A novel type of plastic crystal composite polymer electrolyte has been synthesized by UV-irradiation process successfully. This new polymer electrolyte (denoted as “MB-PCPE”) is composed of a UV-cured trimethylolpropane propoxylate triacrylate (TPPTA) macromer/poly(vinylidene fluoride-co-hexafluoropropylene (P(VDF-HFP)))/Al<sub>2</sub>O<sub>3</sub> nanoparticles and the succinonitrile-mediated plastic crystal electrolyte (1 mol L<sup>-1</sup> lithium bis-trifluoromethane sulphonimide/succinonitrile (LiTFSI/SN)). The unique composition/structure of MB-PCPE brings remarkable improvement in mechanical flexibility and thermal stability. The MB-PCPE has not broken down after 200 cycles in the bending test. The thermogravimetric analytical result shows that no obvious weight loss can be found for the MB-PCPE until above 200 °C. Moreover, the MB-PCPE can display favorable electrochemical performances. The ionic conductivity of MB-PCPE may reach  $1.03 \times 10^{-3} \text{ S cm}^{-1}$  at room temperature and the electrochemical stability window is up to 5.2 V (vs. Li/Li<sup>+</sup>). Furthermore, the MB-PCPE shows outstanding interfacial stability toward lithium metal electrodes. The LiFePO<sub>4</sub>/Li cell assembled with MB-PCPE reveals excellent cycling performance and rate capability, which can exhibit a discharge capacity of 155.4 mAh g<sup>-1</sup> at a current density of 0.1 C after 50 charge–discharge cycles and a discharge capacity of 122.5 mAh g<sup>-1</sup> at a current density of 2 C, respectively. Owing to the beneficial properties, the MB-PCPE is considered to have significant potential applications for rechargeable lithium-ion batteries.

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## 1. Introduction

All-solid-state polymer lithium-ion batteries have received considerable attention with the development of portable electronics, roll-up display devices and wearable electronic instruments [1,2]. Compared to the liquid electrolyte, polymer electrolyte is an attractive alternative due to their favorable properties, which include excellent thermal and electrochemical stabilities, fast ion transport and good flexibility [3,4]. Polymer electrolyte can be divided into two types: the gel polymer electrolyte and dry state electrolyte [5,6]. Gel polymer electrolyte is usually formed by immersing large amount of organic liquid electrolyte into a polymer framework. They exhibit very high ambient temperature ionic conductivities and low rates of safety failure. However, the gel polymer electrolyte has still been suffered several disadvantages, such as worsening of mechanical performances, increasing of reactivity towards the lithium metal electrode and releasing of volatiles [7,8]. The dry state electrolyte is prepared by dissolving lithium salts into a

solid polymer framework as the case in the familiar polyethylene oxide (PEO) membrane. Nevertheless, the ambient temperature ionic conductivities of the dry state electrolyte are very low for practical applications owing to their slower segmental chain mobility in the crystalline regions compared to the amorphous regions [9,10]. Recently, a plastic crystal electrolyte (PCE), which is composed of lithium salts and plastic crystals with solvation capability, have been garnered considerable attentions owing to their unique characteristics of soft matters [11,12]. Plastic crystal is a kind of mesophase formed predominantly by quasi-spherical or disk-like molecules demonstrating rotational and/or orientational disorder while preserving a long-range translational order [13,14]. One representative example is the succinonitrile (SN, NC–CH<sub>2</sub>–CH<sub>2</sub>–CN), which keeps plastic crystalline phase in a temperature range between approximately –40 °C and 60 °C [15]. Due to the presence of non-ionic plastic crystalline phase, SN can be employed as a solid solvent to dissolve lithium salts. SN/lithium salt-based PCE, which exhibits a high ionic conductivity of more than  $10^{-3} \text{ S cm}^{-1}$  at room temperature, has a unique structure of trans-gauche isomerism involving rotation of molecules about the central C–C bonds of SN [16,17]. Unfortunately, the mechanical strength of SN/lithium salt-based PCE is not great enough for large-scale applications.

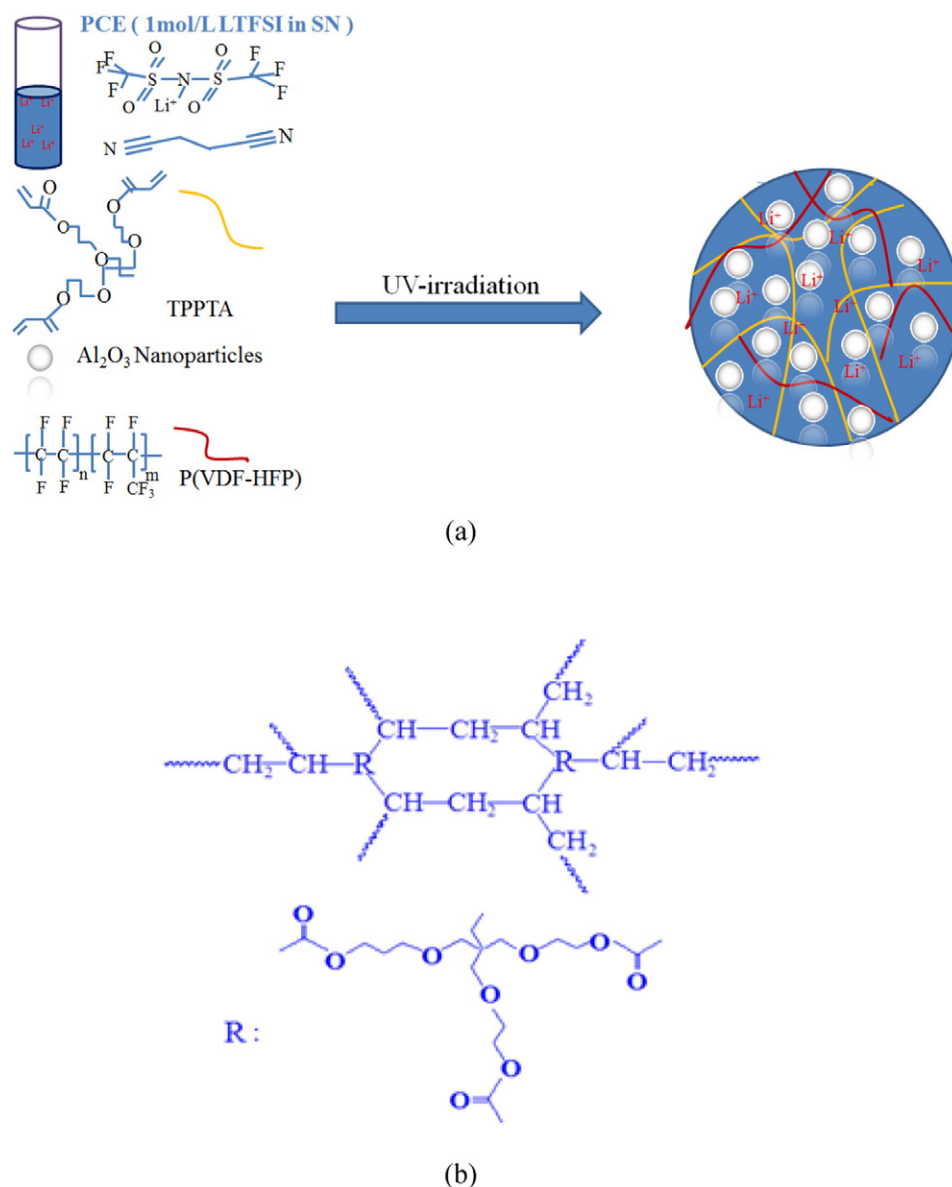
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To improve the mechanical properties of plastic crystal electrolyte, exploiting a plastic crystal composite electrolyte (PCCE) by combination of the PCE with a polymer matrix, which may provide the mechanical framework, is a feasible method [18,19]. In recent years, several research progresses on the PCCE prepared by the ultraviolet (UV)-irradiation method have been reported. H.J. Ha et al. have studied a self-standing plastic crystal composite electrolyte (X-PCCE), wherein UV-cured ethoxylated trimethylolpropane triacrylate (ETPTA) networks are incorporated into a plastic crystal electrolyte (PCE, 1 M lithium bis-trifluoromethanesulphonamide (LiTFSI) in succinonitrile (SN) [20]. H.J. Ha et al. have also developed a UV-cured PCCE based on integration of semi-interpenetrating polymer network (semi-IPN) matrix with a plastic crystal electrolyte (PCE, 1 M LiTFSI in SN). The semi-IPN matrix in the PCCE is composed of a UV-crosslinking polymer network of ETPTA and a high-mechanical-strength polymer of poly(vinylidene fluoride-co-hexafluoropropylene) (P(VDF-HFP)) [21]. K.H. Choi et al. have fabricated a bendable plastic crystal polymer electrolyte (B-PCPE) in flexible lithium-ion batteries. This B-PCPE is composed of a plastic crystal electrolyte (PCE, 1 M LiTFSI in SN) and a UV-cured polymer network of trimethylolpropane propoxylate triacrylate (TPPTA) with a long linear hydrocarbon chain [22]. K.H. Choi et al. have also

developed a deformable safety-reinforced plastic crystal polymer electrolyte (N-PCPE) for high-performance flexible lithium-ion batteries. The N-PCPE is fabricated by combining a plastic crystal polymer electrolyte with a porous polyethylene terephthalate (PET) nonwoven. The three-dimension reticulated plastic crystal polymer electrolyte matrix is formed directly inside the PET nonwoven skeleton by *in-situ* UV-crosslinking of ETPTA monomer under co-presence of the PCE [23]. S.H. Kim et al. have also prepared a shape deformable and thermally stable plastic crystal composite polymer electrolyte (PC-CPE). The PC-CPE is composed of UV-cured ETPTA macromer/closed packed  $\text{Al}_2\text{O}_3$  nanoparticles framework and the succinonitrile-mediated plastic crystal electrolyte (1 M LiTFSI in SN) [24].

In this study, we prepared a new plastic crystal composite polymer electrolyte with high mechanical bendability (referred to as “MB-PCPE”) by an ultraviolet irradiation (UV-curing) process. The MB-PCPE is composed of the plastic crystal electrolyte matrix (providing the ion transport channel) and a composite of UV-cured TPPTA/P(VDF-HFP)/ $\text{Al}_2\text{O}_3$  framework (acting as the mechanical skeleton). The schematic representation illustrating the UV-cured fabrication process and the molecular structure of MB-PCPE after polymerization is shown in Scheme 1. Compared to other plastic-crystal composite electrolyte



**Scheme 1.** Schematic representation illustrating the UV-cured preparation process and chemical structure of MB-PCPE (a). The molecular structure of MB-PCPE after polymerization (b).

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