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## **ACCEPTED MANUSCRIPT**

# Polymeric Ionic liquid based interpenetrating polymer network for all-solid self-standing polyelectrolyte material.

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KEYWORDS.

poly (ionic liquid), interpenetrating polymer network, epoxy-amine, polyelectrolyte.

#### ABSTRACT.

The rise of smart and flexible electrochemical devices requires the design of robust and highly ion conductive separators able to operate on a large temperature range. Polymeric ionic liquids (PILs), having ionic liquid-like species on their backbone, are promising to develop all-solid electrolyte but lead to compromises between ionic conductivity and mechanical properties. We copolymerized an ionic liquid monomer (ILM) with poly(ethylene glycol) dimethacrylate (PEGDM) to give rise to x-PIL networks. They can be used as polyelectrolyte from -46 °C to 220°C. The most ion conducting x-PIL is 80/20 ILM/PEGDM ( $4.8 \times 10^{-5}$  S.cm<sup>-1</sup> at 25 °C). For mechanical improvement, it was associated with an epoxy-amine (EA) network via Interpenetrating Polymer Network (IPN). The IPNs have better hand tear resistance than x-PIL and are rubbery above 5°C with E' at 1 MPa. The EA network plays the role of a reinforcing scaffold without impacting ion conduction of  $10^{-4}$  S.cm<sup>-1</sup>.

#### 1. INTRODUCTION.

lonic liquids ILs are salts in the liquid state at low temperature with very low vapor pressure. The growth of the number of publications on the ILs can be explained by their interest for the development of "green chemistry". They would limit the use of volatile organic compounds in industrial processes [1]. Another obvious field of application is their use as electrolytes in advanced electrochemical devices [2]. Indeed, additionally to their large electrochemical windows, they have two main advantages: a very high concentration of ions and a high ionic mobility at room temperature. Their ionic conductivities can be up to  $10^{-2}$  S.cm<sup>-1</sup>. So, it is possible to envisage a number of applications as electrolytes [3] in rechargeable lithium-ion batteries [4], fuel cells [5–7], solar cells [8,9] or other electroactive devices [10–14].

The development of modern electrochemical devices encompasses by the design of new non-volatile, robust and highly ion conductive solid separators that are necessary to improve the device operation. The use of "polymeric ionic liquids" (PILs), having ionic species covalently bonded with a polymer backbone, appears as a promising approach. It allows combining all beneficial properties of

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