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## Ionogels and ionoelastomers as ionic conducting membranes with healable properties for soft actuators and sensors

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### Abstract:

Ionogels are organic materials consisting of ionic liquids (ILs) immobilized in a polymer network. They combine the mechanical properties of polymer networks with the ionic conductivity, non-volatility, and non-flammability of ILs. Ionoelastomers are elastomers based on crosslinked poly(ionic liquid) (PIL) and have been introduced recently as liquid-free and stretchable ionic conductors. As being ionically conductive, flexible and stretchable, ionogels and ionoelastomers have attracted huge attention in the field of wearable electronics, soft actuators or flexible sensors. However, in the frame of such applications, these materials are usually subjected to repeated deformation, making them susceptible to damage. In these works, we propose to enable the healing ability of these materials by the introduction of dynamic exchangeable bonds, more specifically by the synthesis of associative covalent adaptable networks, also called vitrimers.

First, ionogels are prepared by introducing dynamic  $\beta$ -hydroxy ester linkages. The networks are obtained by the *in-situ* step-growth polymerization between poly(ethyleneglycol) bis(carboxymethylether), trimethylolpropane triglycidyl ether (TMPTTE) as crosslinker and poly(ethyleneglycol) diglycidylether as chain extender via carboxylic acid – epoxy addition reaction. The resulting ionogels are soft, stretchable and conductive, with a tensile strength of 100 kPa, a maximum elongation of 300 % and an ionic conductivity of  $2 \times 4.10^{-4} \text{ S.cm}^{-1}$  at 30°C (with 50 wt. % of IL). The ionogel exhibits strain sensing ability (Gauge factor = 2.8 at 100 % strain), and low hysteresis. (see figure 1). [1] Concerning ionoelastomers, they are obtained in two-steps from a commercially available copolymer of polyepichlorohydrin and poly(ethyleneoxide). The commercial rubber is first modified to introduce allyl-imidazolium cationic side groups with bis(trifluoromethanesulfonyl)imide mobile counter-ions. The resulting linear PIL is then crosslinked through thiol-ene photoaddition thanks to a dithiol crosslinker bearing dynamic boronic ester (see figure 2). The resulting ionoelastomer is soft with a Young modulus of 0.2 MPa, stretchable up to 300% elongation, and exhibits an all-solid state ionic conductivity of  $1.6 \times 10^{-5} \text{ S.cm}^{-1}$  at 30 °C. [2] Both materials demonstrated efficient healing thanks to the dynamic vitrimer chemistry with a full recovery of mechanical and conducting properties. First examples of integrations of these materials in smart polymeric devices will be presented as well.

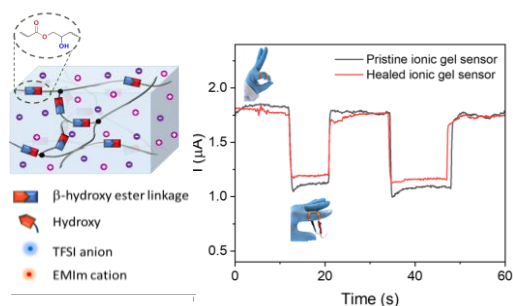


Figure 1: Vitrimer ionogels with strain sensor properties

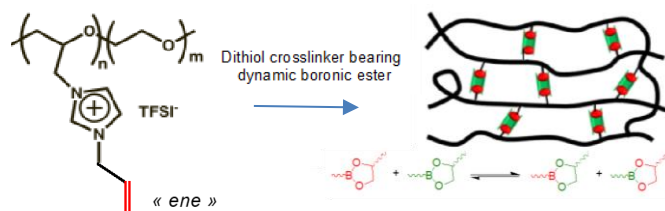


Figure 2: synthesis of the ionoelastomer from PIL

### References :

- [1] K. Bui, G. T.M Nguyen, C. Vancaeyzeele, F. Vidal, X. Hu, C. Wan, C. Plesse, 'Stretchable, healable, and weldable vitrimer ionogel for ionotronic applications', Chemical Engineering Journal 2023, 474, 145533.  
[2]: F. Li, Giao. T. M. Nguyen, C. Vancaeyzeele, F. Vidal, C. Plesse, 'Healable Ionoelastomer Designed from Polymeric Ionic Liquid and Vitrimer Chemistry', ACS Appl. Polym. Mater. 2023, 5, 529–541.