

***From Smart Devices to Healable Ionoelastomers:  
Advances in Ionic Electroactive Polymers and vitrimeric electrolytes.***

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This presentation will start with an overview of the recent developments in smart materials and devices based on ionic electroactive polymers (i-EAPs) that have been conducted at the LPPi. Thus, soft actuators and electrochromic/electroemissive devices currently under development will be presented. These systems function as electrochemical cells where the synergy between electrodes and electrolytes determines overall performance. By utilizing electronic conducting polymers as i-EAPs, we exploit redox-driven transitions that alter electronic conductivity, optical absorbance, and volume, offering key advantages for the resulting devices such as flexibility, lightweight design, and low power consumption.

The second part of the presentation focuses on ionoelastomers, which are based on polymeric ionic liquids (PIL) and have been recently introduced as liquid-free and stretchable ionic conductors [1-3]. In this study, the ionoelastomer is designed as an all-solid electrolyte that has the potential for integration into our devices. The material under study is based on a PIL synthesized from a poly(epichlorohydrin-co-ethylene oxide) copolymer. The PIL was cross-linked via thiol-ene photoaddition using a crosslinker bearing dynamic covalent bonds. This approach resulted in a polymer electrolyte with vitrimer properties. Under optimized conditions, the ionoelastomer is soft (Young's modulus of 0.2 MPa), highly stretchable (up to 300% elongation), and exhibits an ionic conductivity of  $1.6 \cdot 10^{-5} \text{ S}\cdot\text{cm}^{-1}$  at 30 °C. The reported material has allowed obtaining sustainable high-performance polymer materials with high ionic conductivity and by combing the facile processability of thermoplastics with the performance advantages of permanent crosslinked thermosets via vitrimer chemistry.

[1] Kim, H. J.; Chen, B.; Suo, Z.; Hayward, R. C. Ionoelastomer Junctions between Polymer Network of Fixed Anions and Cations. *Science* (80-. ). 2020, 367 (6479), 773–776. <https://doi.org/10.1126/science.aay8467>.

[2] Kim, H. J.; Paquin, L.; Barney, C. W.; So, S.; Chen, B.; Suo, Z.; Crosby, A. J.; Hayward, R. C. Low-Voltage Reversible Electrodeposition of Ionoelastomer Junctions. *Adv. Mater.* 2020, 32 (25). <https://doi.org/10.1002/adma.202000600>.

[3] Ming, X.; Zhang, C.; Cai, J.; Zhu, H.; Zhang, Q.; Zhu, S. Highly Transparent, Stretchable, and Conducting Ionoelastomers Based on Poly(Ionic Liquid)S. *ACS Appl. Mater. Interfaces* 2021, 13 (26), 31102–31110. <https://doi.org/10.1021/acsmi.1c05833>.