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Thermotropic Ionic Liquid Crystals: Structure/ion transport correlation within stimuli-responsive electrolytes for energy

Hélène PUNG¹, Celso Yassuo OKADA-JUNIOR², Mirella SIMOES SANTOS³, Sébastien LIVI², Jannick DUCHET-RUMEAU², Agilio PADUA³, Patrice RANNOU⁴, Manuel MARÉCHAL^{1,*}

¹Univ. Grenoble Alpes, CNRS, CEA, IRIG-SyMMES, 38000 Grenoble, France

²Université de Lyon, CNRS, Univ. Claude Bernard Lyon 1, INSA Lyon, Univ. Jean Monnet, UMR 5223, Ingénierie des Matériaux Polymères, 69621 Lyon, France

³Laboratoire de Chimie, Ecole Normale Supérieure de Lyon, CNRS, Lyon, 69342 France

⁴Univ. Grenoble Alpes, Univ. Savoie Mont-Blanc, CNRS, Grenoble INP, LEPMI, Grenoble, 38000 France

*manuel.marechal@univ-grenoble-alpes.fr

Thermotropic Ionic Liquid Crystals (TILCs)¹ have recently emerged as a smart class of organic electrolytes encoding the unique hierarchical self-assembling properties and stimuli-responsive mosaicity of mesophases featured by thermotropic liquid crystals² with the efficient ion transport of ionic liquids. As a result, TILCs stand out as a promising electrolytic platform for energy storage (batteries³/supercapacitors) and conversion (fuel cells/DSSCs) as they use industry-compatible and simple-to-implement manufacturing processes. The multi-scale correlation relating their structures and (an/cat)ionic transport properties is crucial to pave understanding of their ionic transport. A fundamental question of interest is here the influence of the dynamical mosaicity related to the average size of their self-assembled domains, with or without (w vs. w/o) dynamic grain boundaries. Remarkably, stimuli such as electric⁴ or magnetic field can be leveraged to sample (dis)order till ultimately reaching monodomains (eradication of mosaicity) to reveal inherent (*i.e.* defect-free) features of their ionic transport.

Exploring this research endeavour, in this communication, the case study of an anionic conductive TILC (A-TILC) composed of an imidazolium-based cation with symmetrical *n*-alkyl chains (C₁₈C₁₈Im⁺) and a N(CN)₂⁻ anion featuring a Smectic A (SmA⁵) mesophase (lamellar organization) will be deciphered by combining *state-of-the-art* (SoA) theoretical and experimental approaches. First, coarse-grained simulations derived from Martinelli⁶ will be presented to unravel its set of structural and dynamic descriptive parameters. Second, an unique *operando* experiment combining (synchrotron-based) SoA structural studies (SAXS+WAXS) with ionic transport measurements (probed by EIS) w vs. w/o a tuneable (0 up to 1T) magnetic field (**Fig. 1**) will be introduced, allowing in-depth study of the interplay linking its structure with its ion transport properties. In closing and beyond the case study of an A-TILC encoding a nanoconfined 2D transport within a SmA mesophase, we will illustrate the potential offered by TILCs to impart next generation (conversion/storage) energy devices with self-healing functionalities.

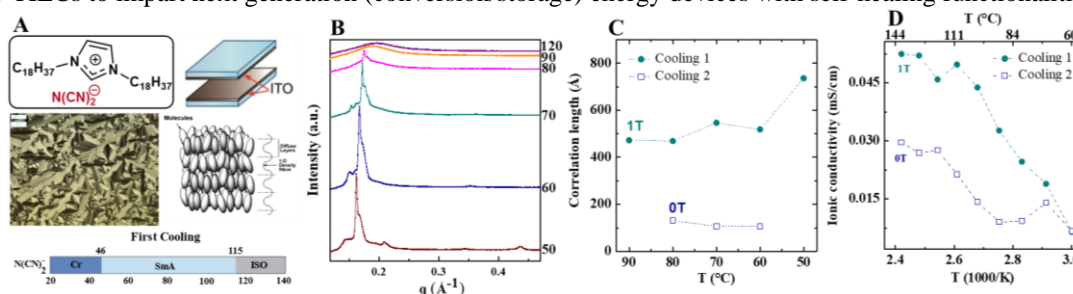


Fig.1. A. Chemical structure of the C₁₈C₁₈Im⁺/N(CN)₂⁻ TILC, LC cell (through-plane configuration) and mesomorphic behaviours (phase transition and texture) of its SmA mesophase. B. 1D SAXS/WAXS scattering profiles (first heating) as a function of temperature. C. vs. D. Temperature dependent evolution of the correlation length (average domain size) and ionic conductivity w vs. w/o 1T magnetic field.

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