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

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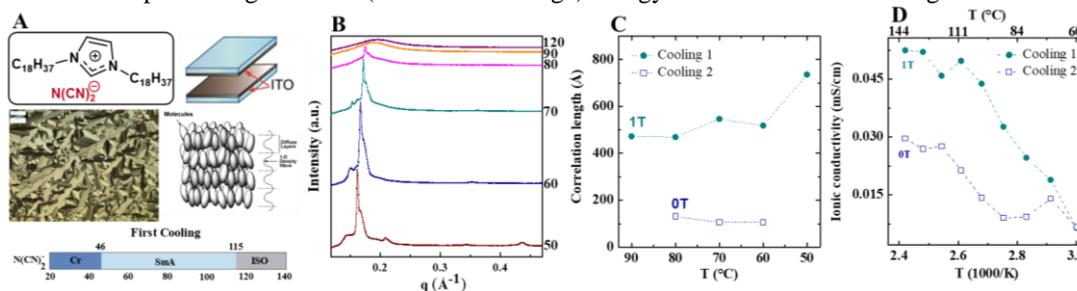
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Thermotropic Ionic Liquid Crystals (TILCs)<sup>1</sup> have recently emerged as a smart class of organic electrolytes encoding the unique hierarchical self-assembling properties and stimuli-responsive mosaicity of mesophases featured by thermotropic liquid crystals<sup>2</sup> with the efficient ion transport of ionic liquids. As a result, TILCs stand out as a promising electrolytic platform for energy storage (batteries<sup>3</sup>/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 electric<sup>4</sup> 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 (C<sub>18</sub>C<sub>18</sub>Im<sup>+</sup>) and a N(CN)<sub>2</sub><sup>-</sup> anion featuring a Smectic A (SmA<sup>5</sup>) mesophase (lamellar organization) will be deciphered by combining *state-of-the-art* (SoA) theoretical and experimental approaches. First, coarse-grained simulations derived from Martinelli<sup>6</sup> 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 C<sub>18</sub>C<sub>18</sub>Im<sup>+</sup>/N(CN)<sub>2</sub><sup>-</sup> 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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**References:** <sup>1</sup>K. Goossens *et al.*, *Chem. Rev.* **2016**, *116*, 4643. DOI: [10.1021/cr400334b](https://doi.org/10.1021/cr400334b) <sup>2</sup>J. Uchida *et al.*, *Adv. Mater.* **2022**, *34*, 2109063. DOI: [10.1002/adma.202109063](https://doi.org/10.1002/adma.202109063) <sup>3</sup>Z. Ahmad *et al.*, *Proc. Natl. Acad. Sci. (USA)* **2020**, *117*, 26672. DOI: [10.1073/pnas.2008841117](https://doi.org/10.1073/pnas.2008841117) <sup>4</sup>H. Shimura *et al.*, *Adv. Mater.* **2009**, *21*, 1591. DOI: [10.1002/adma.200802252](https://doi.org/10.1002/adma.200802252) <sup>5</sup>I.W. Hamley, *Soft Matter* **2022**, *18*, 711. DOI: [10.1039/d1sm01758f](https://doi.org/10.1039/d1sm01758f) <sup>6</sup>G.D. Barbosa. *et al.*, *Macromolecules* **2022**, *55*, 26. DOI: [10.1021/acs.macromol.1c01932](https://doi.org/10.1021/acs.macromol.1c01932)