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Ionic Liquids/Ionic Liquid Crystals for Safe and Sustainable Energy Storage Systems

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Abstract

Ionic liquid crystals are organic salts having synergistic properties of ionic liquids and liquid crystalline materials endowed with non-covalently bound delocalised ion pairs of large organic cations and anions. They can undergo stimulus-responsive anisotropic phase change, followed by enhancement in ionic diffusion and conductivity, which makes them ideal candidates as electrolyte in energy storage systems. Our goal in this chapter is to survey the key developments in the field of ionic liquid crystalline electrolytes and to generate curiosity in the wider research community in tackling challenges in the electrolyte materials for sustainable energy related devices, such as supercapacitors, Li batteries, fuel cells and dye-sensitized solar cells (DSSCs).

Keywords: ionic liquids, ionic liquid crystals, energy storage, supercapacitors, batteries

1. Introduction

Research and development, in the arena of sustainable energy, is receiving overwhelming interest due to the rapid proliferation of portable nano-electronic devices and also evolution in lifestyle. Due to the depletion of petroleum-based energy resources, researchers are turning towards the energy production from wind, solar, tidal, hydro and geothermal energy sources. But these resources are intermittent with time and season. Safe, sustainable and clean strategies for energy storage, such as supercapacitors, batteries, fuel cells, etc. have been explored tremendously in recent years to store energy in a sustainable way. Electrodes and electrolytes are the main components present in the energy storage devices. Electrolytes are the materials through which the transport of ions takes place from anode to cathode and vice versa, during charge-discharge processes. Transport and diffusion of ions through the electrolyte are largely influenced by various material parameters, such as viscosity, porosity and ionic conductivity.
of the electrolytes. Among the presently used liquid electrolytes, ionic liquids (ILs) are delivering excellent performance due to their high ionic conductivity, wide electrochemical stability window, good thermal stability, wide liquidity range, non-volatility and non-toxicity. Being composed of ion pairs bound by non-covalent interactions, they are bestowed with innumerable fascinating properties suitable for various applications [1, 2]. Generally, ionic liquids contain the cationic centres, such as nitrogen, oxygen, sulphur, phosphorous atoms and counter anions, such as halides, hexafluorophosphate (PF$_6^-$), tetrafluoroborate (BF$_4^-$), bis-triflimide [(CF$_3$SO$_2$)$_2$N]$^-$, etc. as shown in Figure 1 [3–10].

![Figure 1. Common cations and anions of ionic liquids.](image)

The structural design of ionic liquids is playing a very important role as it is decisive for controlling the temperature limits, self-assembly and associate functional properties. Self-assembly processes are mainly controlled by the type of cations, anions and side-chains. In ILs, only delocalised charge centres are present which makes the intra-molecular interactions less intense to gift them their exquisite properties. The organic cations and anions of ILs are often stabilised by resonance with highly delocalised charges. Ions with lower symmetry or without symmetry are another major feature of ILs, which restricts the molecular packing into a low-energy crystal structure, ultimately lowering the melting point [11, 12]. Their physical and functional properties could be well tuned, since they are capable for infinite number of
structural variation by changing cations, anions and the substituents [13–17]. They exhibit high
degree of miscibility with a wide array of organic and inorganic materials, since they contain
both organic cations and inorganic anions or vice versa. ILs is found to have higher affinity
towards CO\textsubscript{2} and water vapour. This could be explored in devising gas separators for removing moisture or poisonous gases from the mixtures and also helping in CO\textsubscript{2} sequestration and reducing the greenhouse effect [18–22]. The main criteria for exploiting a compound as an electrolyte for electrochemical energy storage systems are high ionic conductivity, non-volatility, non-flammability, high thermal stability and good electrochemical window. Electrochemical window can be defined as the limiting potential range, in which, the electrolyte is electrochemically stable and is free from any unwanted redox reactions. As the electrochemical window of electrolyte is directly related to the energy density, high EC window is necessary for electrolytes for energy storage systems. In comparison with the commercially used electrolytes, ILs has greater ionic conductivities and better potential windows to draw superior energy storage performances. Unlike aqueous electrolytes with limited potential window less than 1 V, IL-based electrolytes have very high window of 3–4 V, very high ionic conductivity, very good liquidity range and thermal stability for efficient energy storage applications, such as electrochemical supercapacitors, Li-ion batteries, Na- or Mg-batteries, Fuel cells, etc. [23].

Compared to ILs, ionic liquid crystals (ILCs) are receiving importance in flexible electronics as safe and efficient electrolytes. They exhibit synergistic properties of both ionic liquids and liquid crystals. Just like ILs, ILCs are also solely composed of ions bound with ionic interactions in different extents. Liquid crystals are often regarded as the fourth state of matter, with ordering in-between highly rigid crystalline lattices and disordered liquid state. Mesophase formed between solid and liquid phases exhibit impressive properties due to positional order and flow properties. Rotating isotropic spherical molecules are responsible for the plastic crystal formation, whereas, the non-covalent interactions are behind the anisotropic behaviour of liquid crystals. Ionic interactions are often responsible for strong crystalline ordering, whereas, other non-covalent interactions like hydrogen bonding, van der Waals forces and layer-by-layer self-assembly are responsible for liquid crystalline mesophase formation. In addition to the cationic and anionic centres, long alkyl side-chains and functional groups are present in ILCs for modulating the mesophase self-assembly. They are the real smart materials for realising multi-functional applications, such as stimuli-responsive conductors for energy storage devices, electrochromic supercapacitors, Li-ion batteries, Na- or Mg-batteries, Fuel cells, etc. Stimuli-responsive phase transition of ILCs enables to modulate the direction of ionic conduction in them as shown in Figure 2. Based on the order and texture of mesogens, mesophases exhibited by ILCs are mainly of two types: nematic phase and smectic phase [24]. Mesogenic molecules tend to align along the molecular axis in the nematic phase (1D ordered). Smectic phase is more ordered where the molecules are arranged in layers (2-dimensionally ordered). Based on the extent of ordering of mesogens within the layers, smectic phase is subdivided into Sm A, Sm B, Sm C, Sm D, Sm F, Sm G, etc. Sm A is the least ordered one, among all the smectic mesophases [25–28]. Tschierske defined the LC state as a condensed matter state, in which, there is orientational and/or positional long-range order in at least one direction and no fixed position for individual molecules [25]. Columnar and cubic phases are other major mesophases observed in ionic liquid crystals. Based on the lattice in which columns are arranged, columnar phase is divided into hexagonal columnar (Col\textsubscript{h}), rectangular (Col\textsubscript{r}) and oblique
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