Application of alkoxysilyl-functionalized imidazolium-based ionic liquids as quasi-solid electrolytes in electrochromic devices

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Ionic liquids (ILs) have become important as electrolytes in various iono-optic and electrochemical devices due to their high ionic conductivity, thermal stability, and wide electrochemical potential window [1]. However, despite their low vapor pressure, they are still liquid at room temperature. This inspired the investigation of their solidification either by using small weight gelators, via addition of solid nanoparticles, or through incorporation of the IL in polymer matrices. Another possibility is the synthesis of ionic liquids with polymerizable cations or anions (vinyl, acryl groups, etc.) [1] that assure the formation of polymeric electrolytes of suitable mechanical and rheological properties in-situ in an electrochemical device. Following this approach, various alkoxysilyl-functionalized imidazolium based ionic liquids [2, 3], i.e. new compounds belonging to the Class II of organic-inorganic hybrids were prepared.

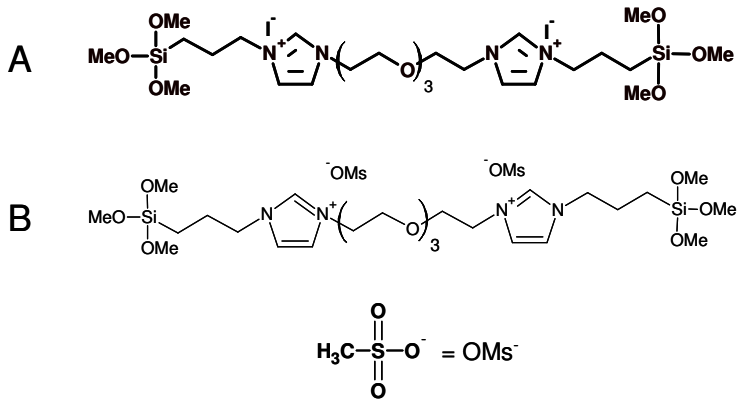

Figure 1. Structures of two bis end-capped ionic liquids: A) iodide IL and B) mesylate IL.

Different types of alkoxysilyl ILs were synthesized, either with iodide or mesylate anions, as bis end-capped or single end-capped compounds, and with various lengths of ethylene oxide chains to influence the solubility of lithium salts. As solvolysis catalysts, various organic acids were evaluated such as acetic or formic acid. The structural evolution of the electrolytes during the sol-gel process was followed using ^{29}Si NMR, ^{1}H NMR, IR, and Raman spectroscopy.

Electrochemical impedance spectroscopy (EIS) measurements revealed that the conductivity of electrolytes can reach the order of magnitude of 0.001 S/cm when an appropriate amount of non-reactive alkyl-functionalized ionic liquid is added to the bis end-capped precursor. The obtained gels were elastic and characterized by good gluing properties. It was found that a mixture of the bis end-capped iodide ionic liquid and 1-methyl-3-propyl imidazolium iodide in a molar ratio of 1:10 sustained at least 15.000 coloring and bleaching cycles when employed in a hybrid electrochromic device based on Pt and WO_3 electrodes. In this device, iodine was added to the electrolyte to form triiodide/iodide redox pairs that can react on the Pt electrode, while lithium ions are intercalated/deintercalated into/from the WO_3 film. A similar procedure was used for the preparation of electrolytes on the basis of mesylate ILs that were applied in a battery-type electrochromic device composed of two films with intercalation properties.

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