Self-assembled materials consisting of organic molecules have attracted a great deal of attention for application in energy devices. In particular, liquid crystals with well-defined channels for the transport of ions are prospective as electrolytes. Recently, we applied layered 2-dimensional liquid-crystalline (LC) ion conductors in lithium batteries and dye-sensitized solar cells (DSSCs). In the present work, LC ion conductors composed of carbonate functionalized rod-shaped mesogens and ionic liquids (ILs) have been developed and applied in DSSCs. The liquid crystals show layered smectic A phases in wide temperature ranges, up to over 110 °C. Remarkably, the DSSCs based on these electrolytes show significant improvement in power conversion efficiency on heating and can continue to operate until over 100 °C. Furthermore, the LC-DSSCs show high open-circuit voltage compared to IL based devices. These results promote liquid crystals as a new class of electrolytes for solar cells capable of efficient conversion of light into electricity at elevated temperatures.

**Introduction**

Liquid-crystalline (LC) complex nanostructures formed by self-assembly of organic molecules have attracted a great deal of attention as functional materials.\(^\text{[1,2]}\) Particularly, liquid crystals with well-defined channels for the transport of ions are prospective as electrolytes for energy devices. Recently, we applied 2-dimensional LC ion conductors in lithium batteries.\(^\text{[3]}\) In the present work, LC ion conductors consisting of carbonate functionalized mesogens (1) and ionic liquid (2) (Figure 1) have been developed and applied as electrolytes in dye-sensitized solar cells (DSSCs).\(^\text{[4]}\)

**Figure 1:** Schematic illustration of the 2-dimensional liquid-crystalline electrolyte.
Mixtures composed of compound 1 and ionic liquid (IL) 2 in different ratios was prepared. The LC behavior of the mixtures was evaluated by polarized optical microscopy (POM), differential scanning calorimetry and x-ray diffraction. These LC mixtures were applied as electrolytes in dye-sensitized solar cells (DSSCs). POM was used to observe the organization of the liquid crystals inside the devices. The mass transport properties of the electrolytes, device performances, and the charge reactions at the electrode interfaces of the solar cell devices were evaluated as a function of temperature. A comparative study between LC-based and IL-based devices was conducted.

The mixtures show 2D layered nanostructures in wide temperature ranges, up to over 110 °C (Figure 2a). POM observation revealed that the liquid crystal show polydomainal alignment inside the devices (Figure 2b). The best performing device contained the electrolyte mixture composed of 40 mol% 1 and 60 mol% 2 (1/2-(60)). Interestingly, the 1/2-(60)-based DSSCs exhibit high open-circuit voltage ($V_{oc}$) compared to the IL(2)-based devices. Furthermore, the LC-DSSCs show significant improvement in power conversion efficiency (PCE) on heating, while the IL-devices show a decrease in PCE (Figure 2c). The beneficial temperature behavior of the LC-DSSCs was found to be related to the electron life-time ($\tau$) in the TiO$_2$ electrode. The 1/2-(60)-based devices show higher $\tau$ than the IL-devices over the studied temperature interval (Figure 2d). The increase in $\tau$ negatively shifts the quasi-fermi level of the TiO$_2$ which consequently leads to higher $V_{oc}$ and improved device performance at elevated temperatures. These results promote liquid crystals as a new class of electrolytes for development of photovoltaic devices capable of efficient conversion of light into electricity at elevated temperatures.

Figure 2: a) Phase diagram of electrolyte mixtures. b) Polarized optical micrograph of 1/2-(60) in a solar cell. c) Power conversion efficiency (PCE) as a function of temperature for 1/2-(60)-DSSCs (red dots) and 2-DSSCs (black square). d) Electron life-time ($\tau$) of the TiO$_2$ of 1/2-(60)-DSSCs (red dots) and 2-DSSCs (black squares).