Mapping the electrochemical double layer of ionic liquids as fuel cell electrolytes by atomic force microscopy

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Proton-conducting ionic liquids are promising candidates as next-generation electrolytes for intermediate-temperature polymer electrolyte membrane fuel cells (PEMFC) and an operation temperature of 100–130 °C, which is highly beneficial for future applications in electric vehicles based on the use renewable energy. Designing a suitable ionic liquid with sufficient proton solubility and conductivity as well as a high stability is demanding, since the properties of ionic liquids are significantly different from that of an aqueous solution [1]. Ionic liquids are composed entirely by ions leading to a superposition of Coulomb interaction between anions and cations with steric effects related to the spatial orientation of the rather large organic ions. In consequence, a complex bulk structure exhibiting a domain-like character evolves.

The presence of the catalytic Pt electrode, where the redox reactions in the fuel cell take place, introduces a further complexity in the system. Due to the presence of a surface charge upon electrode polarization, additional Coulomb forces are present rearranging the structure of the ionic liquid in the electrochemical double layer [2].

In order to investigate the properties of this double layer by a direct method, we employ atomic force microscopy in an electrochemical cell. By recording force-separation curves on a single crystalline Pt electrode in the model ionic liquid diethylmethylammonium trifluoromethanesulfonate, [Dema][TfO], we map the interface structure of the electrode-electrolyte interface with sub-nanometre resolution.

Depending on the polarization of the electrode, a dense layered structure of alternating anion and cation layers can be identified as characteristic jumps in the force-separation curves when approaching a Si tip to the Pt surface in the ionic liquid. This interface structure depends not only on the polarization of the Pt electrode but also gets diluted upon humidification. This shows that the presence of water strongly affects the double layer structure, that will in turn have a significant impact on the electrochemical performance of a fuel cell based on ionic liquids.

Literature:

- [1] K. Wippermann, J. Wackerl, W. Lehnert, B. Huber, and C. Korte, Journal of The Electrochemical Society **163**, F25 (2016).
- [2] C. Rodenbücher, K. Wippermann, and C. Korte, Applied Sciences 9, 2207 (2019).