

NEW INSIGHTS INTO SEI FORMATION ON GRAPHITE IN ALKYL CARBONATES AND DINITRILES BASED SOLVENTS

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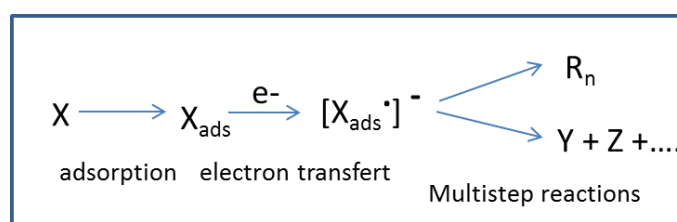
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In order to increase the energy density of Li-ion batteries for portable applications the use of higher cell voltages or larger specific capacities is required. Most anodes material, like graphite, silicon or tin are working out of the electrochemical stability range of the commonly used electrolytes. Then, stable cycling cannot be achieved without the formation of a stable and Li^+ conducting solid electrolyte interphase (SEI). Using high voltage cathode ($>5\text{V}$) cannot be achieved with alkyl carbonates (AC) as electrolyte solvent as they are not enough stable. Alternative solvents which are more resistant to oxidation, like dinitriles (DN), may be used instead of AC but the formation of a stable SEI on the negative electrode has to be re-considered.

The simpler manner to stabilize the anode/electrolyte interface is to use additives such as vinyl or fluorinated derivative of ethylene carbonate (EC) which have already identified to be efficient for increasing batteries performances and cycle life.

The mechanism of action of SEI former compounds is not completely understood today. Nevertheless some recent works [1-4] are able to provide new insights based on the scheme reported below:



R_n : insoluble polymeric network
 Y, Z : small molecules and ions, oligomers

where X represents the SEI former additive. When the applied potential is driven to sufficiently low value, X is reduced in a multistep process which leads to a polymeric 3D network (R_n) and by products (Y, Z). Some of the byproducts have been already identified in solution or at the electrode/separators surface by mean GC-MS analysis, IR spectrometry and XPS analysis but nothing is known at this time about the polymer molecular weight.

In this presentation, it will be shown that SEI former additives are often strongly adsorbed at the electrode interface before being reduced and this explain why they are efficient even at very low concentration and in the presence of various solvents including DN.

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- [4] G. Gachot, P. Ribiere, D. Mathiron, S. Grugeon, M. Armand, J.-B. Leriche, S. Pilard and S. Laruelle, Anal. Chem., 83 (2011) 478–485.