THE CONTRIBUTION OF THE DIELECTRIC SPECTROSCOPY

TO THE STUDY OF THE ELECTRODE MATERIALS

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The improvement of battery performance requires the rationale optimization of the composite electrode. The developments of new experimental techniques as well as methodologies are needed to understand the relationships between the composition, the architecture and the performance of composite electrodes. The fruitful contribution of Broadband Dielectric Spectroscopy (BDS) to study hierarchical materials applied to lithium ion and lithium metal batteries electrodes have been previously shown ¹⁻⁴. The results demonstrate that the broadband dielectric spectroscopy technique is very sensitive to the different scales of the electrode architecture involved in the electronic transport, from interatomic distances to macroscopic sizes, as well as to the morphology at these scales, coarse or fine distribution of the constituents. When the frequency increases, different kinds of polarizations appear from macroscopic sizes to interatomic distances and give rise to dielectric relaxations in the following order: (a) space-charge polarization (low-frequency range) due to the interface sample/current collector; (b) polarization of clusters (or agglomerates) of particles (micronic scale); (c) polarization of particles due to the existence of resistive junctions between them; (d) electronic transfers (at nanometric or at interatomic scale). The BDS measurement was up to now ex situ measurement, on dry electrode. They provide a fundamental insight into the conduction properties at all scales of the materials before being integrated in a real battery. An innovative device (measurement cell) has been developed to make synchronized BDS measurements and electrochemical cycling: the frequency range is about $10^2 - 10^{10}$ Hz. In this work, data acquisitions were made on dry electrodes (Active material / Carbon black / PVdF) and then on the same electrode wetted with an electrolyte. Short- and long-range motions of ions are evidenced in the low-frequency region. At higher frequencies, the study shows for the first time the influence of the ions of the electrolyte on the transfer of the electronic charges (and conversely) at the micronic and nanometric scales ⁴. The new device opens thus important prospects to determine the evolutions of the multi-scales electrical properties during electrochemical cycling.

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