



PhD thesis proposal (CEA Saclay / IMN) – October 2018

Title : Ageing processes in Lithium-ion batteries studied by radiolysis

Commercialized since 1991, lithium-ion batteries (LIBs) are becoming a flagship technology able to power from microchips to large-scale application markets such as electric vehicles. LIBs performance issues mainly involve the chemical decomposition of the electrodes and electrolyte. These degradations are specific to the combination of compounds used (active material/solvents/Li salt). Ageing studies are usually performed on different electrode/electrolyte systems in full cell batteries. However, these studies are lengthy, costly, most often purely qualitative, and thus inappropriate for batch tests. A fast screening method is thus essential to establish which the optimal system is. We have recently discovered and proven that radiolysis (i.e., the chemical reactivity induced by the interaction between matter and ionizing radiation) is a powerful tool for a short time (within minutes) identification of the degradation products as compared to several months when cycling is used. Based on these results, the aim of this PhD thesis is to extend the radiolysis approach to:

- screen several combinations of electrolyte/active materials to identify the most robust ones;
- study carefully the interfacial processes (electrolyte/electrode) with negative and positive electrodes for the most interesting systems previously identified.

This work will be performed both at NIMBE laboratory (CEA-CNRS, Gif-sur-Yvette) and at IMN institute (Univ. Nantes-CNRS) since it is funded by CEA and by Region Pays de la Loire. A company (Solvionic, <http://fr.solvionic.com/>) will be also involved in this particular research activity.

The first part of this PhD thesis will be dedicated to a **screening** of different electrolytes (carbonate- and ionic liquid-based) as well as electrolyte/active materials combinations in order to improve the safety, stability and cyclability of the future LIBs. This screening will be done by gas chromatography (GC): it will easily enable **identifying and quantifying the main gases** produced upon irradiation; the most stable systems producing the lowest gas amounts. Moreover, all the various tools provided by mass spectrometry (MS) will be used to identify and quantify degradation products formed both in the gaseous and liquid phases (**EI-MS, GC-MS and FT-ICR MS**) and then, to propose reaction mechanisms. For the most interesting electrolytes identified, picosecond pulse radiolysis will be performed at the ELYSE facility in Université Paris Saclay (Orsay).

The second part will be dedicated to the study of the **interphases created between an electrode material and the electrolyte**. The most interesting electrolytes identified previously will be chosen and associated with typical negative electrode materials such as carbon and also the very challenging silicon. Newly developed nanostructured materials will be studied in order to compare their properties with conventional electrode materials. Most robust electrolytes will also be studied when associated with high voltage positive electrode materials. Multiple spectroscopic techniques **XPS, solid-state NMR, infrared spectroscopy and EELS** will be used characterize the interphase created. In situ microscopy in a newly acquired corrected transmission electron microscope (Themis Z) will also be performed.

Applicant profile: you have obtained a Master degree (M2 or equivalent) in Materials science, Analytical, Physical- Chemistry or equivalent. You have a good knowledge of interaction of light and/or electrons with mater and associated characterization techniques (as given above). You have a strong interest in understanding physical and chemical phenomena and are able to deal with multiple environments and teams.

Contact: CV + application letter + least 2 year marks to:

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