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Accelerated ageing of electrolytes in Lithium-ion batteries: the point of view of a radiation chemist

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Since the early nineties, the rechargeable Li-ion battery (LIB) technology has dominated the electronic market. These batteries have become essential components in portable electronic applications. Since then, ageing processes are a growing global concern, essentially for their applications in electric and hybrid vehicles.

Ageing phenomena occurring in diethylcarbonate (DEC), DEC/LiPF₆ propylene carbonate (PC) and PC/LiPF₆ solutions, selected as model systems, have been studied using gamma- and pulse- radiolysis as a tool to generate similar species as the ones occurring in electrolysis of Li-ion batteries and to mimic the processes (Fig. 1) [1-3]. We prove that similar results were obtained in the ageing of an electrochemical cell filled with the same model solution [2]. This highlights the interest of the radiolysis approach in the field of ageing of electrolytes. Radiolysis has indeed the following major advantages: i) ageing processes are strongly accelerated (minutes/hours as compared to weeks and months in conventional battery studies); ii) time-resolved experiments are possible, enabling to study the system on multiple temporal scales (from picoseconds to minutes and days) [2]. This allows measuring rate constants, and writing very detailed reaction mechanisms; iii) the possibility to study the reactivity of each solvent without/with the salt (of course studies without the salt has no sense in the battery field!), leading to a very accurate understanding of the behavior of the system; iv) the possibility to perform a quick screening of many electrolytes, to identify rapidly the most promising ones. All these points will be discussed.

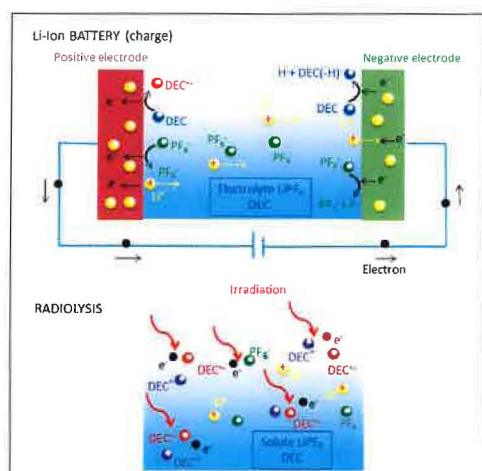


Fig. 1. Scheme comparing primary electron transfers at the electrodes in the electrolytic charge/ageing processes of a Li-ion battery (with molar DEC/ LiPF₆ solution as a model electrolyte) (top), and after ionization in the bulk in the radiolytic process with the same medium (bottom).

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