Ageing processes in lithium-ion batteries deciphered thanks to radiolysis

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Among many energy storage devices, Lithium ion batteries (LIBs) are efficient power sources used for various applications including mobile microelectronics i.e. phones and laptops. However, aging phenomena causing loss of performance are not yet fully understood. Moreover, these phenomena are a crucial issue related to providing safe and stable batteries. LIBs are usually composed of an electrolyte, a lithium metal oxide cathode and an anode where the active material is graphite. We have recently shown that radiolysis is a powerful tool to simulate the degradation of the electrolyte in short time: minutes/hours instead of weeks/months by electrolysis [1-3]. Radiolysis also enables experiments at the picosecond time scale thus giving access to reaction mechanisms [2-6]. Indeed, we have shown that the highly reactive species created in the irradiated solution [2,3] are the same as the ones obtained during the charging of a LIB using similar solvents. We have also evidenced that linear and cyclical carbonates exhibit very different types of reactivity [2,4,5] and studied the particular features of a mixture of linear and cyclical carbonate [6].

During the first cycles of the battery, the negative electrode reacts with the electrolyte leading to the formation of a solid interphase between the anode and the electrolyte (solid electrolyte interphase, SEI). We show that an artificial SEI can be produced by radiolysis. Compared to the case of radiolysis on electrolyte, we observe the same species as well as similar degradation mechanisms. Interestingly the composition of the formed artificial SEI depends on the carbonaceous material. The SEI obtained at the surface of graphite is composed of Li carbonate, oxalate and oligomers of poly(ethylene oxide) while the SEI formed at the surface of amorphous carbon nanoparticles contains Li salts such as Li_2CO_3. Therefore, radiolysis is a promising tool to rapidly simulate the ageing behavior of different anode materials and facilitate the development of next generation batteries.

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