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▶ **To cite this version:**

Marin Puget, Viacheslav Shcherbakov, Sergey Denisov, P. Moreau, Jean-Pierre Dognon, et al.. Reaction Mechanisms of Fluoroethylene Carbonate Degradation, an Additive of Lithium-Ion Batteries, Unraveled by Radiation Chemistry. EMIRUM 2021, Jan 2021, Palaiseau, France. cea-03126694

HAL Id: cea-03126694
https://cea.hal.science/cea-03126694

Submitted on 1 Feb 2021

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Reaction Mechanisms of Fluoroethylene Carbonate Degradation, an Addi- tive of Lithium-Ion Batteries, Unraveled by Radiation Chemistry

Marin Puget[1], Viacheslav Shcherbakov[2], Sergey Denisov[2], Philippe Moreau[3], Jean- Pierre Dognon[1], Mehran Mostafavi[2] and Sophie Le Caër[1]

[1] NIMBE, UMR 3685 CEA, CNRS, Université Paris-Saclay, CEA Saclay F-91191 Gif-sur-Yvette Cedex, France.

[2] Institut de Chimie-Physique/ELYSE, UMR 8000 CNRS/Université Paris Saclay, F-91405 Orsay Ce- dex, France.

[3] Institut des Matériaux Jean Rouxel, IMN, Université de Nantes, CNRS, 2 rue de la Houssinière, BP 32229 F-44322 Nantes Cedex 3, France.

sophie.le-caer@cea.fr

Abstract

Numerous additives are used in electrolytes of lithium-ion batteries, especially for the formation of ef- ficient solid electrolyte interphase at the surface of the electrodes. It is, therefore, necessary to elucidate the degradation processes of these compounds since it directly affects the lifetime of the battery. These mechanisms can be obtained through radiolysis. In this work, we investigated the degradation mecha- nisms induced by irradiation in fluoroethylene carbonate (FEC), a cyclic carbonate, which is an additive commonly used in lithium-ion batteries. The first reaction steps were studied by pulse radiolysis. At long timescales, the radiolytic yields of produced gases (H_2, CO, and CO_2) were quantified. Pulse radi- olysis experiments evidenced the formation of the $FEC^{\bullet-}$ radical anion, characterized by an absorption band centered ca. 430 nm. The radical anion is not detected when FEC is solubilized in other solvents: ethanol, diethylcarbonate, etc. This radical is indeed stabilized in neat FEC, whereas the ring opens to form more stable radical anions when FEC is a solute in other solvents, as confirmed by calculations. A multi-species deconvolution of the spectrum measured in pure FEC revealed a small absorption band centered around 560 nm, attributed to the solvated electron, decaying in ca. 100 ps. In neat FEC, excess electrons primarily undergo attachment compared to solvation. Together with gas chromatography cou- pled to mass spectrometry measurements, all these observations have allowed us to propose a reaction scheme for both the oxidizing and reducing pathways at stake in irradiated FEC. This work gives clues for the reaction mechanisms undergone by FEC present in electrolytes of lithium-ion batteries and evi- dences that the nature of the primary species formed in FEC depends on the amount of FEC in the solution.