

# Simulation of Electrolyte Molecule Degradation on Electrode Surfaces: A Comparison of Methods - Molecular Dynamics and Genetic Algorithm

**BOUDER Gaëlle,<sup>A</sup> CARBONNIERE Philippe,<sup>A</sup>**

A) IPREM UMR5254, Institut des Sciences Analytiques et de Physico-chimie pour l'Environnement et des Matériaux, Université de Pau et des Pays de l'Adour, E2S UPPA, CNRS, Technopôle Hélioparc, 2 Av. du Président Pierre Angot, 64053, Pau, France

[gaelle.bouder@univ-pau.fr](mailto:gaelle.bouder@univ-pau.fr)

All-solid-state batteries (ASSBs) with a polymer electrolyte have emerged as a promising solution to address the growing demand for efficient energy storage systems and reduce overall energy consumption<sup>1</sup>. These batteries utilize a solid-state electrolyte composed of a polymer material, offering numerous advantages over traditional liquid electrolytes<sup>2</sup>. However, the utilization of a lithium metal anode gives rise to several additional challenges<sup>3</sup>. Namely, the intricate relationship between electrolyte degradation mechanisms and the growth of the electrode-electrolyte interface. This investigation aims to describe such a mechanism and predict the possibility of avoiding it in order to extend battery's lifespan.

In the present work, ab initio molecular dynamics (AIMD) simulations and Genetic Algorithm (GA) using density functional theory (DFT)<sup>4</sup> are employed to investigate the degradation of a polymer electrolyte on distinct lithium metal anode surfaces. To this purpose, we realized different systems with isolated molecules of the electrolyte on Li-metal: one molecule of FEC (solvent) and one molecule of LiFSI (lithium salt) (figure 1). Then, we investigated the decomposition of these molecules, with and without the protective layers consisting of inorganic materials (Li<sub>2</sub>O, LiF) and carbon-based 2D materials (Graphyne, Graphdiyne). The current study is devoted to comparing two methods in terms of computation time and the quality of results.

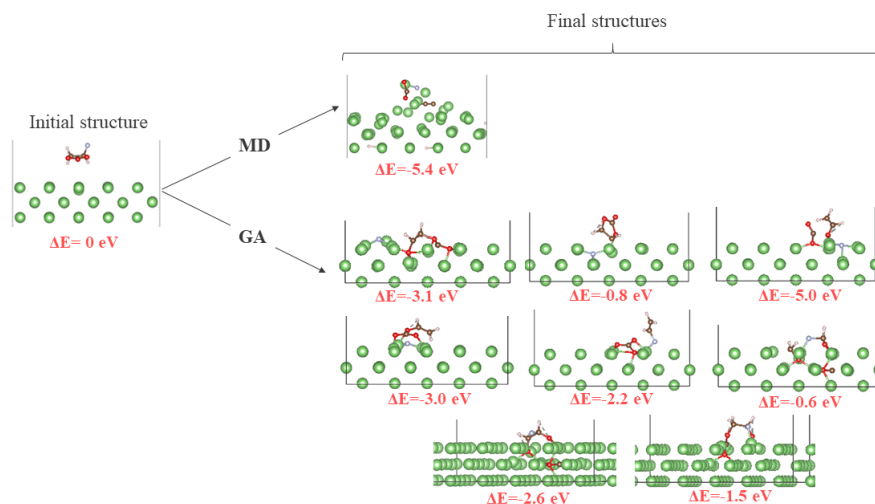


Figure 1. Decomposition products for FEC on Li surface using two methods: MD (Molecular Dynamic) and GA (Genetic Algorithm)

**Keywords:** Density functional theory; Li-metal battery; Electrodes protection; Solid electrolyte interphase.

<sup>1</sup> A. C. Luntz, J. Voss, and K. Reutner, *J. Phys. Chem. Lett.* 6 (2015) 4599–4604

<sup>2</sup> C. Zheng, J. Wu, L. Zhang, and H. Wang, *J. Mater. Chem. C* 11 (2023) 344–363

<sup>3</sup> Y.-Y. Sun, Q. Zhang, L. Yan, T.-B. Wang, and P.-Y. Hou, *Chemical Engineering Journal* 437 (2022) 135179

<sup>4</sup> P. Hohenberg, and W. Kohn, *Phys. Rev.* 136 (1964) B864