

The faculty of Engineering of the Vrije Universiteit Brussel invites you to attend the public defense leading to the degree of

DOCTOR OF ENGINEERING SCIENCES

of **Seyed Hamidreza Beheshti**

The public defense will take place on **Friday 19th April 2024 at 4:00 pm** in room **I.0.02** (Building I, VUB Main Campus)

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Meeting ID: 347 804 988 496

Passcode: YXpYy4

**EFFECTS OF SUBSTITUTED CARBONYL COMPOUNDS IN CYCLIC
FORMATION OF SOLID-ELECTROLYTE INTERPHASE IN LI-ION
BATTERIES**

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Abstract of the PhD research

Lithium-ion battery (LIB) has been a disruptive technology offering new possibilities to the energy sector, especially in the growing market of electric vehicles. The formation of a decent Solid-Electrolyte Interphase (SEI), the passivation layer formed on the anode particles during the initial cycles, ameliorates LIB specifications in terms of capacity, power output, cycle life, and safety resulting in more efficient cells. SEI features are dependent on electrolyte content as this complex layer originates from the electrolyte decomposition products. Accordingly reformulating the electrolyte by introducing interphase-improving additives is followed as a facile and cost-effective approach to reach a performant SEI. Despite all the advantages, conventional SEI additives are toxic materials that multiplying their share into the enormous yet growing number of produced cells arises an environmental issue hence investigating new compounds to mitigate such concerns is required.

Regarding the anodic SEI formation mechanisms, a slight modification in the molecular structure of the electrolyte components causes fundamental changes in their electro-reduction behaviour, where adjusting the electronic arrangement within the molecule could be an effective approach to improve the functionality of additives. Also, since the elemental content of the SEI forming agent determines the interphase composition, controlling the substituents could affect the stability of the reduction products. Additionally, since the electro-reduction reactions occur via the generation of radical ions, charge stabilizing features are another aspect that should be considered in additive studies.

Considering the flexible functionalization of the carbonyl group which makes it interesting chemistry in molecular design, this work is devoted to understanding the performance of its derivatives in the SEI formation and LIB cyclability. Also, given the environmental footprints of the SEI improving agents, we have targeted to study the performance of nontoxic chemistries. The selected structures cover carboxylic acids, esters, and acyl halides so the effects of these functional groups could be compared as a novel approach. In addition, changes caused by the vicinity of the phenyl group to the polymerizable C=C bond are tracked to understand its impact on the graphite SEI formation process.

To track the role of additives in SEI evolution, first, we have comprehensively reviewed the current understanding of the SEI formation processes of different molecular structures. Having focused on carbonyl compounds, the potential stability of the selected molecules is studied both by computational modelling (DFT) and electrochemical analysis (Voltammetry). In the next step, the cyclic behaviour of the electrolytes is comparatively assessed both in half-cell and full-cell configurations followed by investigating the impacts of C-rate and temperature as SEI-affecting parameters. Additionally, employing electrochemical impedance spectroscopy (EIS) complemented with surface morphology characterization (SEM) and compositional analysis (XPS), the chemical and electrochemical evolution of the interphase and the correlations between them have been investigated.

Results have depicted the evolution process of the formed SEI layers that gradually transforms the interphase toward a sectional film with higher content of inorganic species close to the graphite electrode suggesting the corresponding mechanisms. It is shown that systematic structural adjustments within the SEI forming additives affect the interfacial chemistry, leading to changes in the composition and impedance of the layer where molecular tuning ameliorates the cyclability of the electrolyte leading to a higher capacity retention of the LIB cell and better resilience against operating conditions.