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  

**Lieven Bekaert**

The public defense will take place on **Tuesday 12th March 2024 at 2:00 pm** in the **Green Room** (U-Residence, VUB Main Campus)

**Entrance at the back of the building**

To join the digital defense, please click [here](#)
Meeting ID: 358 347 883 319
Passcode: iDQSKj

**TOWARDS A STABLE METAL ELECTRODE / SOLID-STATE ELECTROLYTE INTERFACE IN RECHARGEABLE LI-ION AND NA-ION BATTERIES THROUGH MULTISCALE MODELLING**

**BOARD OF EXAMINERS**

Dr. Xinhua Zhu  
Prof. dr. ir. Marijke Huysmans  
Prof. dr. ir. Dimitrios Angelis  
Prof. dr. Alejandro Franco  
Prof. dr. Jean-François Gohy  
Dr. Julia Contreras Garcia

**PROMOTORS**

Prof. dr. ir. Annick Hubin  
Prof. dr. Frank De Proft  
Prof. dr. ir. Mesfin Haile Mamme
Abstract of the PhD research

Rechargeable batteries are at the basis of the modern digital society. Since the commercial introduction of the lithium-ion battery in the 1990s, they have become widely used in phones, cars, and recently also in stationary storage to store energy from renewable resources. At present, a continuous improvement of their performance, safety, cost, and sustainability is ongoing. To enhance the safety, conventional liquid electrolytes are being replaced by solid-state electrolytes. To increase the energy-density, the use of metal electrodes is gaining popularity. Furthermore, sodium is being studied as a contender to lithium, owing to its greater abundance and lower cost. With the introduction of new electrode and electrolyte materials, understanding the role of the interface between these materials is vital. An unstable interface is a significant cause of poor battery performance and lifespan. It is therefore essential that this is studied in depth.

In this work, the interface between the solid-state electrolyte and the negative electrode was studied using a combined computational chemistry and experimental analysis approach. *Ab initio* (Density Functional Theory; *Ab initio* Molecular Dynamics) and neural network potential molecular dynamics were used to investigate the interactions and reactions at the interface region. These techniques were combined with newly developed data analysis techniques to analyse combined kinetic and thermodynamic aspects. These results were then corroborated by experiments using X-ray Photoelectron Spectroscopy. The influence of the high reactivity of lithium and sodium metal electrodes on a bio-based solid-polymer electrolyte and a sulphide inorganic solid electrolyte was investigated. Following these insights, a new electrolyte material was discovered which exhibits a superior stability against the sodium metal electrode. Finally, design criteria for materials to achieve a high stability towards reactive metal electrodes are proposed based on the principle of "kinetics engineering".

Overall, this Ph.D. contributes to a better understanding of the negative electrode / solid-state electrolyte interface in rechargeable solid-state lithium and sodium batteries. It also provides new approaches for controlling and limiting the reactivity which can be used to improve battery performance and lifespan.