Secteur des Sciences et Technologies

Invitation à la soutenance publique de thèse de

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Master of Applied Science in Mechanical Engineering

Pour l'obtention du grade de Docteur en sciences de l'ingénieur et technologie

« Ab initio study of lithium dynamics in silicon-based materials for battery applications »

qui se déroulera
le vendredi 20 mars 2020 à 16h
Auditoire SUD 08
Place Croix du Sud
1348 Louvain-la-Neuve

Jury members :

Prof. Xavier Gonze (UCLouvain), supervisor
Prof. Bernard Nysten (UCLouvain), chairperson
Prof. Gian-Marco Rignanese (UCLouvain), secretary
Prof. Jean-François Gohy (UCLouvain)
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Silicon-based materials are promising candidates to replace commercial graphite-based anodes in lithium ion batteries. Silicon-based anodes can possess 10 times more capacity than graphite, but with a humongous volume expansion of 300 %. At the atomic scale, the rate of lithium migration plays a key role in controlling the volume expansion. Thus, understanding the lithium atom migration and the structural evolution of Si atoms at atomic scale is essential to mitigate volume expansion and achieve maximum capacity. In this thesis, we approached this problem from two ends: 1) Studying diffusion of lithium in bulk silicon and lithiated LiSi from first-principles methods within the transition state theory framework, and 2) Analysing delithiation dynamics through first-principles followed by an empirical potential to study large supercells. Our approach provides a better agreement with the experimental data as compared to previous experimental or theoretical data with useful insight on the diffusion mechanism. The effects of quantum tunneling and anharmonicity on lithium diffusion have also been estimated for the first time and are moderate for Si and negligible for the LiSi case. A novel first-principles-based approach has been developed to study step-by-step delithiation of lithium atoms, which can capture the change in the local environment of silicon during delithiation. The analysis of the local environment of delithiated Si suggests that most of the delithiated silicon atoms (75%) returns back to the tetrahedral coordination. The characterization of delithiated silicon obtained by empirical potential show strong resemblance with amorphous Si.