DEPARTMENT OF CHEMICAL AND BIOMOLECULAR ENGINEERING

PhD Dissertation Defense

Sungwon Park

Theoretical Investigation on the Sodiation Mechanism of Electrode Materials for Sodium-Ion Batteries

Sodium-ion batteries (SIBs) have recently become an attractive alternative to lithium-ion batteries (LIBs) because of the low cost and natural abundance of Na resources. However, the commercialization of SIBs is hampered by the limited choice of suitable electrode materials providing high-energy density and high-rate capability. The proposed research in this dissertation aims to develop a better understanding of the sodiation process of promising electrode materials using density functional theory (DFT) calculations. Moreover, based on the improved understanding, we attempt to guide the rational design of electrode materials for SIBs.

The first main study is on investigating the possibility of using phosphorus (P)-doped and oxidized P-doped graphene as an anode in SIBs. An emphasis is placed on elucidating the fundamental chemical properties of increasing Na adsorption on P- and oxidized P-doped graphene. Our results suggest that the Na capacity could reach 511 mAh/g with the P- and oxidized P-doped graphene which exceeds that of hard carbon anode in SIBs. Moreover, the high mobility of electron and Na atoms on P- and oxidized P-doped graphene indicates its potential to achieve good rate performance.

The second part of this dissertation is devoted to examining the sodiation mechanisms of selenium (Se), which is an attractive candidate for cathode in SIBs. We first examine the incorporation and dynamic behavior of Na atoms in crystalline Se; followed by assessing the sodiation-induced changes in the energetics, atomic structure, and electronic/mechanical properties of crystalline and amorphous Na-Se alloys. Next, we identify the sodiation behavior of Se-graphene, representing Se with carbonaceous (C) host materials, with a particular interest in the interface region. Our calculations demonstrate higher mobility of interfacial Na atoms along the graphene surface in comparison to the bulk phase, thus contributing toward high-performance cathodes with fast charge/discharge rates. Lastly, the charge transport mechanism of sodium selenide (Na_2Se), the fully sodiated terminal state of Se as a cathode, is investigated. We examine the contribution of ionic and electronic defects to the conductivity of Na_2Se. Our results suggest that negatively charged Na vacancy is the main charge carrier in Na_2Se primarily due to its high concentration and mobility.

Location: CAMP 372

Date: December 7, 2022 (Wed)

Time: 12:30 PM

Advisor:	Prof. Eunsu Paek
Committee members:	Prof. Elizabeth Podlaha-Murphy
	Prof. Sitaraman Krishnan
	Prof. Taeyoung Kim
	Prof. Yang Yang