**Voltage Dependence of Ni-Li Electrode on its Composition and Structure: A Density Functional Theory Study**

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Recently a new prototype all-solid-state memory device has been fabricated, which consists of Li electrode, Li3PO4, and another electrode such as Au and Ni [1]. The device has a few states with different open-circuit voltages, which can switch using an external applied bias. Though the switching has been demonstrated, its mechanism and the atomic structures of different states have not been well understood yet except that the movement of Li atoms near the electrode-Li3PO4 interface must play a crucial role. In the present study, using the density functional theory calculations, we examine the dependence of electrode open-circuit voltage on its composition and structure to understand the switching mechanism of this novel device.

In this presentation, we focus on the Ni electrode, because Ni does not form alloy with Li, which is expected to make the interface structure simpler. As the first step, we consider models without Li3PO4, more specifically, the Ni(111) slab and Ni Σ5(210) grain boundary structure. The open-circuit voltage of Ni electrode is estimated by the method which has been widely adopted in previous studies on lithium ion batteries [2]. Our calculation results show that sites inside the Ni electrode and at the grain boundary are energetically unfavorable for a Li atom. On the other hand, Li atoms are energetically stable in some cases when they are on the surface of Ni electrode. The open-circuit voltage of Ni electrode decreases as the surface coverage of Li atoms increases. These results suggest that the atomic structures of Ni electrode with different Li surface coverage are likely to correspond to the states with different open-circuit voltages.

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