IMPACT OF LITHIATION ON SI-ANODE/BINDERS INTERFACE FOR NEXT GENERATION LITHIUM IONS BATTERIES

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To achieve higher energy densities in lithium-ion batteries (LiBs), it is imperative to replace intercalation chemistry with anode materials capable of electrochemically alloying with lithium. Silicon emerges as a very promising candidate; however, its adoption is hindered by intrinsic conductivity issues and volumetric expansion challenges[1,2]. Addressing these concerns, the use of polymeric binders has been proposed[3,4] to uphold the structural integrity of the anode. Numerous research endeavors have focused on enhancing the mechanical, elastic, electrical, and ionic properties of binders to be used with silicon anodes[1,3,5,6]. Anyway, a crucial aspect in merging structural and chemical insights lies in the investigation of silicon/polymer interfaces[7]. Our study centers on recently proposed binders for Si anode: polyaniline (PANI), polyaniline functionalized with boronic acid groups (B(OH_2)-PANI), polyvinylidene fluoride (β-PVDF) and a co-polymer binder incorporating poly[vinyl alcohol] (PVA). This presentation aims to deepen and clarify the mechanisms governing the adhesion properties of these polymers on the Si (111) and Si (110) surfaces looking in particular to the role played by the presence of Li inside the anode. Employing first-principles calculations based on density functional theory, we explore the structural and electronic properties, along with the energetics, of PANI, B(OH2)-PANI), β-PVDF and PVA monomers[8] adsorbed on Si surfaces both before and after lithiation (see Fig.1). Our findings reveal that the co-absorption of two monomers enhances adsorption energy, consequently improving the adhesion properties of polymers on the different Si facets but the inclusion of an increasing amount of Li can influence both the anchoring mechanism and the reactivity of the binders. For this reason, we will discuss the structural evolution and corresponding electronic properties in relation to Li concentration.

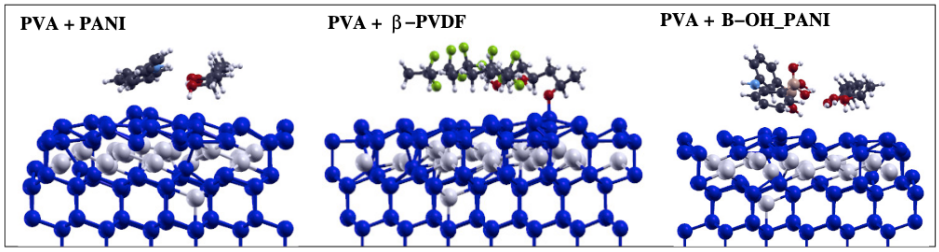

Figure 1 Optimized Li-Si-111 surface with co-binders. Si and Li atoms are presented with blue and grey balls.

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