Correlation between volume expansion and electrode density of Si-Alloy@Graphite composite anode: In-situ Dilatometry Study

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Recently, Si-based anode materials, including alloys, have been intensively studied to replace low-capacity commercial carbonaceous materials (e.g., graphite). However, they are hindered by their low electronic conductivities and huge volume expansion upon the lithiation/de-lithiation process. For the practical application of Si, Si-graphite blending materials were introduced to compensate for the limitation of Si via an easy and simple blending approach [1]. Nonetheless, when it comes to considering volumetric energy, Si blending with graphite composite electrode swelling is one of the most critical factors to be addressed. To achieve high volumetric energy density, the electrode is calendared under high pressure, leading to a decrease in the electrode porosity. Notably, the porosity of the electrode affects electrolyte penetration, capacity fading, rate capability, side reactions caused by the SEI layer and lithium plating, and mechanical degradation due to the charging stress [2,3]. Thus, studying the relationship between electrode density and volumetric energy by controlling electrode swelling is necessary to achieve high energy density and stable electrochemical properties because, without densification of the electrode, the volumetric energy density of the Si@Gr composite electrode is inferior.

The current research investigates the influence of electrode density on electrode swelling of graphite-blended Si-alloy anode via *in situ* measurement of the electrode swelling with electrochemical dilatometry. To study the swelling behavior of the electrode at various electrode densities, single-layer pouch cells were proposed with the same electrodes: NCM523 cathode vs Si-alloy@G anode. Volumetric energy densities were estimated depending on the electrode swelling at various electrode densities. The amorphous Si-alloy was synthesized using a high-output and mass-producible melt-spinning process; then, the as-prepared ribbon is annealed at a certain temperature (by adopting recrystallization phenomena) to homogeneously crystallize and achieved nanosized-Si (n-Si) embedded in an inactive alloy matrix. The crystal structure, microstructure, and grain growth of Si-based alloy were systematically studied via high-temperature X-ray diffraction (HT-XRD) and focused ion beam combined scanning transmission electron microscopy (FIB-STEM).

The results exhibit a tendency that an increasing the high electrode density provokes higher electrode expansion and contraction as well. The expansion/contraction of the Si-alloy@G electrode depending on the electrode density is given in Figure 1. These tendencies indicate that the porosity of the electrode has a significant effect on the electrode swelling.

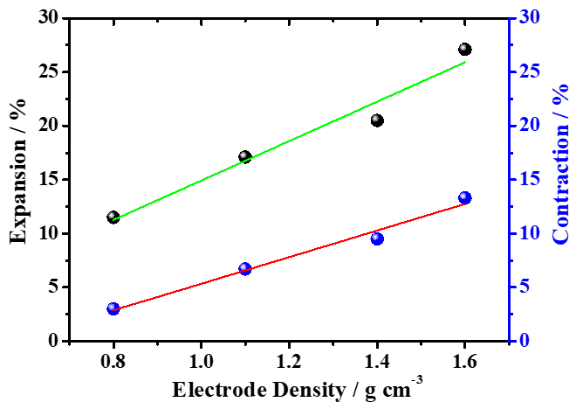

Figure 1. The electrode expansion/contraction versus electrode density in Si-alloy@G electrode.

The results of the study provide an understanding of the importance of optimizing the electrode density to control the volume expansion and achieve a high volumetric energy density of Si blended graphite composite anodes.

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