Prolonged Cycling Life of Si@Graphite Composite Anodes Enabling with a Polymeric Artificial SEI Protective Layer

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There are several advantages including higher specific capacity (higher energy density) and low cost materials when Silicon-on-Graphite composite (Si@Gr) utilizing as anode materials for lithium-ion batteries (LIBs). However, there are several major challenges hindering the development of Si-based anodes, such as the pulverization of Si particles upon (de)lithiation due to the large changes in volume, which can lead to mechanical failure of the electrode, and the low electronic conductivity of Si. The volume changes also lead to the breakage and reformation of the solid-electrolyte-interphase (SEI), causing active lithium losses, electrolyte consumption and building-up of electrolyte decomposition products, resulting in rapid capacity fading and deterioration of the reaction kinetics. Additionally, long cycling stability is another key challenge for commercialization of Si@Gr composites. Herein, to solve these problems, we have developed a multifunctional polymeric artificial solid-electrolyte-interphase (A-SEI) protective layer with thickness in nanometers that was facilely coated on the Si@Gr/Carbon particles by a simple and scalable solution-based method. The A-SEI is composed of crosslinking sulfonated chitosan (SCS) with high degree of sulfonation as main polymer matrix and glutaraldehyde as the crosslinker. The across-linked polymer is insoluble in water and possesses high toughness. The SCS coated Si@Gr/Carbon anodes showed a high specific capacity reaching over 600 mAh g−1. Compared with the anode made of the pristine Si@Gr/Carbon, significantly improved cycling life from 350 to 1000 cycles are achieved with the SCS coated Si@Gr/Carbon anode in Li metal cells. In addition, a significantly improved cycling stability of the modified anodes could also be demonstrated in NCM622||Si@Gr/Carbon-SCS full-cells. The outstanding electrochemical performances can be attributed not only to the A-SEI function of the SCS polymer coating but also the coating layer firmly attach Si nanoparticles and stabilizes the particles. Furthermore, the SCS polymer layer can facilitate the diffusion of Li+ ions between the electrolytes and the Si-graphite composite material due to its cation (Li+)-selective nature. The protective layer can also provide a cushion effect to buffer the large volume change of Si and graphite during the charge/discharge process, which results in an improved Coulombic efficiency by reducing active lithium losses and electrolyte consumption. The simple and scalable polymer coating process by SCS and prolonged cycling life of the resulted electrodes make SCS polymer an attractive coating material for various functional anode and cathode materials of LIBs.