Modeling large patterned deflection during lithiation of micro-structured silicon

The application of silicon (Si) as potential anode material in Li-ion batteries provides a more than nine-fold increase in gravimetric storage capacity compared to conventional graphite anodes. However, full lithiation of Si induces the volume to increase by approximately 300%. Such enormous volume expansion causes large mechanical stress, resulting in non-elastic deformation and crack formation. This ultimately leads to anode failure and strong decrease in cycle life. This problem can be resolved by making use of structured anodes with small dimensions. Particularly honeycomb-shaped microstructures turned out to be beneficial in this respect. In the present paper, finite element modeling was applied to describe the experimentally observed mechanical deformation of honeycomb-structured Si anodes upon lithiation. A close agreement between simulated and experimentally observed shape changes is observed in all cases. The predictive ability of the model was further exploited by investigating alternative geometries, such as square-based microstructure. Strikingly, dimension and pattern optimization shows that the stress levels can be reduced even below the yield strength, while maintaining the footprint-area-specific storage capacity of the microstructures. The pure elastic deformation is highly beneficial for the fatigue resistance of optimized silicon structures. The obtained results are directly applicable for other (de)lithiating materials, such as mixed ionic-electronic conductors (MIEC) widely applied in Li-ion and future Na-ion batteries.