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Numerical modelling of coated silicon nanoparticles during lithiation and core-shell carbon coating optimization.

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Lithium ion batteries (LIBs) using silicon as the anode active material have high energy density and are used in many electronic equipment. A major issue for improving the performance of LIBs is understanding their degradation mechanisms that lead to capacity fade. In this work, we consider that the anode is composed of spherical nanoparticles of silicon in a soft electrolyte media. We are interested in the mechanical behaviour of a single nanoparticle without interaction with the others.

Experimental results [1, 2] indicate that three main phenomena occur during the lithiation of silicon nanoparticles. First, the formation of an advancing lithiation front separating two phases: a pure silicon core and a silicon lithium alloy outer shell. Second, large volume transformation of about 300 % and finally, particle fracture.

In order to take into account those phenomena, semi-analytical and finite element mechanochemistry models were established. The lithiation is treated similarly to a thermomechanical problem, where the lithium concentration drives the differential swelling within the particle. The semi-analytical model is an extension of the composite model of a sphere subjected to a radial loading in the case of elasto-viscoplastic constituents [3]. The finite element model takes into account large deformation via the logarithmic strain framework. The solution in both cases shows that the lithiation front and the viscoplastic deformation of the outer shell are essential ingredients in modelling the lithiation. Moreover, the viscoplastic deformation relaxes the significant internal stresses, induced by the swelling of the silicon lithium alloy, which leads to the formation of a residual tangential traction of the shell. This traction is likely to cause the nanoparticle fracture.

Some experiments show that mitigation of nanoparticle mechanical failure can be achieved by using a carbon coating that has several benefits, such as stress alleviation and swelling restriction. In this work, we analyse the mechanical impact of coating and calculate its optimal thickness using the linear fracture mechanics considering different flaw sizes and geometries.

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