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Pair Distribution Function Analysis and Solid State NMR Studies of Silicon Electrodes for Lithium Ion Batteries: Understanding the (De)lithiation Mechanisms

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The crystalline-to-amorphous phase transition that occurs on electrochemical Li insertion into crystalline Si, during the first discharge, hinders attempts to link the structure with electrochemical performance. We apply a combination of local structure probes, in situ and ex situ 7Li nuclear magnetic resonance (NMR) studies, and pair distribution function (PDF) analysis of X-ray data to investigate the changes in short-range order that occur during the initial charge and discharge cycles. The distinct electrochemical profiles observed subsequent to the first discharge have been shown to be associated with the formation of distinct amorphous lithiated silicide structures. The first process seen on the second discharge is associated with the lithiation of the amorphous Si, forming small clusters. These clusters are broken in the second process to form isolated silicon anions. The (de)lithiation model helps explain the hysteresis and the steps in the electrochemical profile observed during the lithiation and delithiation of silicon. At deep discharge states a highly reactive lithium excess Li15Si4 phase is detected by in situ NMR which undergoes a self-discharge process in electrolyte.