

Towards high energy density silicon-based anodes in all-solid-state lithium batteries

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All-solid-state batteries are expected to provide a solution for the next-generation energy storage applications. The conventional Li-ion batteries consist of flammable organic liquid electrolytes. In order to overcome this safety issue, it is suggested to replace the organic liquid electrolyte by a solid one. In that respect, argyrodite $\text{Li}_6\text{PS}_5\text{Cl}$ is a promising candidate due to its high ionic conductivity and relatively low grain boundary resistance. The main scientific challenge is the interface instability of the solid electrolyte [1] and thus argyrodite towards the active electrode materials, which leads to several parasitic reactions and thereby increasing the overall impedance of the battery[2].

Since argyrodite suffers from a severe interface instability vs metallic lithium as electrode material, silicon was suggested as a promising alternative to replace lithium, still maintaining a high theoretical energy density of 3579 mAhg^{-1} . In combination with a liquid electrolyte the continuous exposure of fresh Si to the electrolyte leads to continuous decomposition of the electrolyte and isolation of the active material. However, in combination with a solid electrolyte the possible decomposition products are immobilized, potentially being an advantage of the Si solid system. Nevertheless, the alloying reaction is accompanied by large volume changes resulting in pulverization of the silicon particles over cycling, which challenges the mechanical stability of the Si solid electrolyte mixture, and thus leading to an irreversible capacity fading.

In the present work, we aim to understand the chemical and electrochemical mechanisms occurring at the interface between the argyrodite $\text{Li}_6\text{PS}_5\text{Cl}$ solid electrolyte and the silicon particles. In this work we assess the stability of $\text{Li}_6\text{PS}_5\text{Cl}$ against pristine silicon compared to pre-lithiated silicon. Characterization of the decomposition products formed on the electrode-electrolyte interface during electrochemical processes was achieved by using solid state NMR. In order to identify eventual crystalline phases present of both pristine and cycled electrodes, powder XRD patterns were collected and interpreted.

References:

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