Operando TEM Studies of the Structural Evolution of All-Solid-State Li-ion Battery

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Conventional lithium-ion batteries (LiB)\cite{1} are the most efficient energy storage devices that triggered the transformation of our lifestyle into a digital nomad with the revolution of portable electronics. Inorganic solid-state electrolytes (SSE) represent a promising alternative due to their nonflammable nature, broader temperature operating range and larger electrochemical window. Therefore, all-solid-state-batteries (ASSB) made an important step toward new-generation in electrochemical energy storage systems\cite{2}. However, several limitations still impact the performances of ASSBs such as SSE ionic conductivity, chemical evolution of SSE/active-materials interface, lithium dendrite growth, grain boundary conductivity and solid/solid interfacial resistance\cite{3}.

To get a better insight into the limiting parameters of the performances of the ASSBs, a better quantification of the relationship between the structural and electrochemical properties is strongly required. In this study we propose an approach to carry out Operando TEM measurements\cite{4} to study the structural and chemical modifications while operating the ASSB inside a TEM, focusing on the multiple solid/solid interfaces. For this study, several ASSBs will be investigated using different combinations of materials with specific SSE such as: Li$_{1.5}$Al$_{0.5}$Ge$_{1.5}$(PO$_4$)$_3$\cite{5}, Li$_6$PS$_5$Cl, LiBH$_4$ and Li$_3$PO$_4$.

A nanobattery is obtained using a FIB milling workstation and electrical contacts are realized on a microchip in order to cycle the ASSB inside of the TEM. The microchip with the nanobattery is characterized during the cycling using a holder which allows heating and biasing and information about the microstructure, the degree of oxidation and chemical composition are obtained.

\begin{enumerate}
\item N. Recham \textit{et al.}, “A 3.6 V lithium-based fluorosulphate insertion positive electrode for lithium-ion batteries,” \textit{Nat. Mater.}, vol. 9, no. 1, pp. 68–74, 2009.
\item Y. Nomura \textit{et al.}, “Quantitative Operando Visualization of Electrochemical Reactions and Li Ions in All-Solid-State Batteries by STEM-EELS with Hyperspectral Image Analyses,” 2018.
\end{enumerate}