Li$_2$S particle size influence on the first charge working mechanism of Li$_2$S-based Li-ion batteries

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With their high theoretical energy density (~2600 Wh.kg$^{-1}$), lithium/sulfur (Li/S) batteries are highly promising, but these systems are still poorly understood due to the complex mechanisms/equilibria involved. Replacing S$_8$ by Li$_2$S as the active material allows the use of safer negative electrode, like silicon, instead of lithium metal. S$_8$ and Li$_2$S have different conductivity and solubility properties, resulting in a profoundly changed activation process during the first cycle. Particularly, during the first charge a high polarization and a lack of reproducibility between tests are observed [1] (Figure 1a). Differences observed between raw Li$_2$S material (micron-sized) and that electrochemically produced in a battery (nano-sized) may indicate that the electrochemical process depends on the particle size [2].

Then the major focus of the presented work is to deepen the understanding of the Li$_2$S material charge mechanism, and more precisely to characterize the effect of the initial Li$_2$S particle size. To do so, Li$_2$S nanoparticles were synthesized according to two ways: a liquid path synthesis [3] and a dissolution in ethanol, allowing Li$_2$S nanoparticles/carbon composites to be made [4]. Electrochemical tests show that starting with Li$_2$S nanoparticles could effectively suppress the high initial polarization (Figure 1b). Then X-Ray Diffraction (XRD) and Electrochemical Impedance Spectroscopy both applied via an operando mode will be presented in order to interpret this observation.

Figure 1: (a) Typical voltage profiles of two first charges of a Li$_2$S-based cell [1] and (b) comparison of voltage profiles between micro and nano-sized Li$_2$S particles-based cell.