

Li₂S particle size influence on the first charge working mechanism of Li₂S-based Li-ion batteries

Alice Robba^{a,b}, Renaud Bouchet^{a,c}, Céline Barchasz^b, Jean-François Colin^b, Fannie Alloin^{a,c}

^a Université Grenoble Alpes, LEPMI, F-38000 Grenoble, France.

^b CEA, LITEN, 17 rue des Martyrs, 38054 Grenoble, France.

^c CNRS, LEPMI, F-38000 Grenoble, France.

email address of the presenting author: alice.robba@lepmi.grenoble-inp.fr

With their high theoretical energy density ($\sim 2600 \text{ Wh.kg}^{-1}$), lithium/sulfur (Li/S) batteries are highly promising, but these systems are still poorly understood due to the complex mechanisms/equilibria involved. Replacing S₈ by Li₂S as the active material allows the use of safer negative electrode, like silicon, instead of lithium metal. S₈ and Li₂S 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 Li₂S 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 Li₂S material charge mechanism, and more precisely to characterize the effect of the initial Li₂S particle size. To do so, Li₂S nanoparticles were synthesized according to two ways: a liquid path synthesis [3] and a dissolution in ethanol, allowing Li₂S nanoparticles/carbon composites to be made [4]. Electrochemical tests show that starting with Li₂S 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.

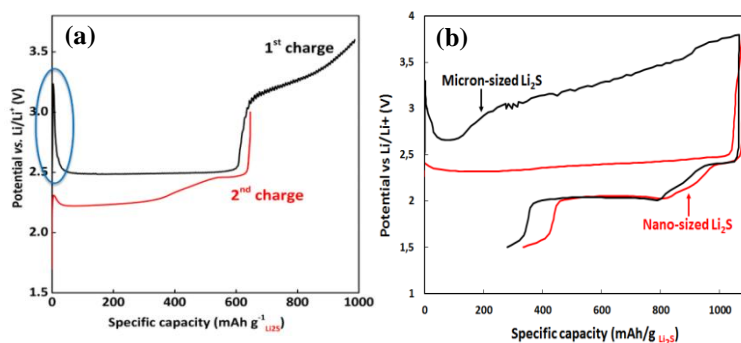


Figure 1: (a) Typical voltage profiles of two first charges of a Li₂S-based cell [1] and (b) comparison of voltage profiles between micro and nano-sized Li₂S particles-based cell.

- [1] S. Waluś, C. Barchasz, R. Bouchet, J.-F. Martin, J.-C. Leprêtre, and F. Alloin, *Electrochim. Acta* 180 (2015) 178–186.
- [2] S. Waluś, C. Barchasz, R. Bouchet, J.-C. Leprêtre, J.-F. Colin, J.-F. Martin, E. Elkaïm, C. Baehtz, and F. Alloin, *Adv. Energy Mater.* (2015).
- [3] G. J.A, K. Wong, and S. Jick, *J.C.S Chem. Comm* (1978) 6–7.
- [4] F. Wu, H. Kim, A. Magasinski, J. T. Lee, H.-T. Lin, and G. Yushin, *Adv. Energy Mater.* 4 (2014).