

Understanding the Battery Degradation Mechanism in All-solid-state Batteries via *In-situ* SEM

Neelam Yadav^{1,2}, Nicolas Folastre,^{1,2} Micakel Bolomont^{1,2}, Arash Jamali³, Mathieu Morcrette^{1,2} and Carine Davoisne^{1,2}

¹ Laboratoire de Réactivité et Chimie des Solides (LRCS), Université de Picardie Jules Verne, Amiens Cedex, France.

² Réseau sur le Stockage Electrochimique de l'Energie (RS2E), France.

³ Plateforme de Microscopie Electronique, Université de Picardie Jules Verne.

neelam.yadav@u-picardie.fr, carine.davoisne@u-picardie.fr

Batteries catching fire while charging have made headlines in recent years. One of the reasons for this is that conventional batteries use liquid electrolytes. This is among the highly cited reasons for the growing interest in solid-state batteries, *i.e.* safety [1]. The other reason is the usage of metallic Lithium as an anode, which will help enhance the gravimetric density of the battery. But all-solid-state batteries (ASSB) is not devoid of problems. To make solid-state batteries a reality, it's important to understand the process responsible for failure in ASSB. 1. During the cell assembly [2] of all-solid-state batteries, parameters such as pressure, temperature, etc. will influence the battery cycling. 2. Also, during battery operation, operating parameters [3,4] will influence battery cycling. They both impact cycling performance leading to battery degradation.

Among different techniques being used to understand the fundamentals of battery degradation, scanning electron microscopy (SEM) offers a good compromise in terms of size and resolution of observation with the possibility to perform studies with a good spatial resolution. By combining the imaging with chemical analyses by X-ray energy dispersive spectroscopy (EDX), a complete survey of morphological and chemical modification is possible. A homemade airtight ASSB cell for air-sensitive has been designed to perform *in-situ* studies in real-time in SEM. The cell has been tested to obtain reliable electrochemistry. The SEM chamber has been modified, to provide a path for the cable inlet and outlet. An *ex-situ* cycling (outside SEM) is first carried out with this cell. Later, similar cycling is carried out inside the SEM to obtain first-hand information and monitor ASSB cycling. This study has been coupled with other techniques, *ex-situ* SEM, XRD, Raman.

Two different types of sulphide based Solid electrolytes have been studied here, namely b-Li₃PS₄ and Argyrodite (Li₆PS₅Cl). A comparative study has been carried out on the failure processes for these two SE's. Though these two SE's are classified under the same category of sulphide based SE's. The former, b-Li₃PS₄ show's huge plating, faster dendrite formation, crack propagation and eventually cell failure when compared to Argyrodite as seen in fig1 below. The role and importance of particles of SE are also highlighted in this study. The *in-situ* SEM study here has been coupled with EIS studies to better understand the electrochemical processes responsible for the degradation of the battery. The observations from this study will help us to design our future ASSB with improved performances.

References:

- [1] L Shen *et al*, *Small* **16** (2020), p. 1.
 [2] L Frenck *et al*, *Front. Energy Res.* **7** (2019).
 [3] Y Wang, T Liu and J Kumar, *ACS Appl. Mater. Interfaces* **12** (2020), p. 34771.
 [4] KN Wood *et al*. *ACS Cent. Sci.* **2** (2016), p. 790.

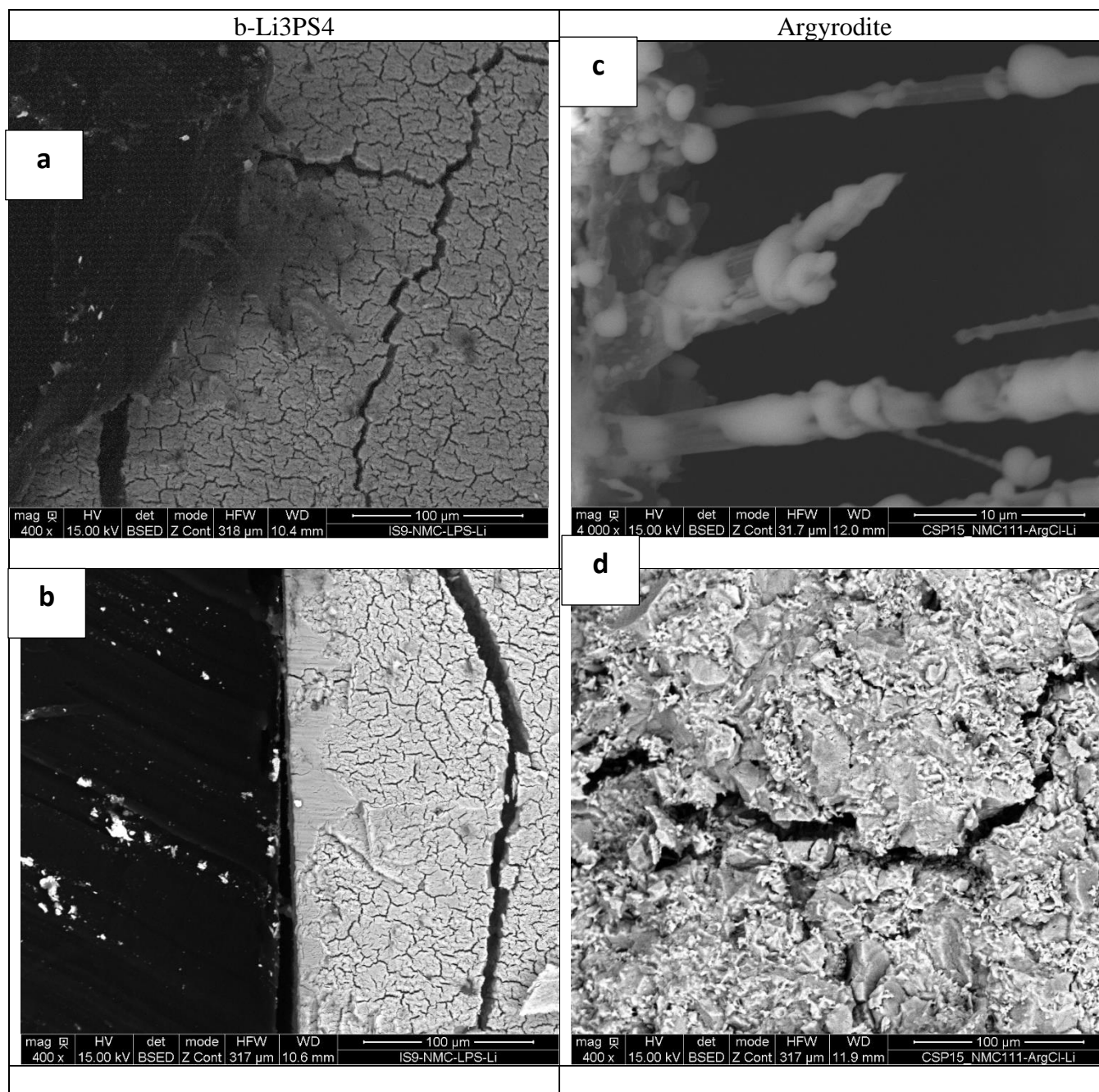


Figure 1. a) and b) plating, cracking and Dendrite formation in Li_3PS_4 ; c) and d) plating and cracks in Argyrodite.