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In-Situ Confocal Microscopy to Analyze Volume Expansion of Solid State Microbatteries

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Abstract Text:

The rise of embedded objects put great interests in miniaturized power supply solutions. Solid State Microbatteries (SSM) are the most promising, as they can safely store high energy and power density. SSM have the advantage of being all-solid (no liquid leakage and flammability), to have low self-discharge and good cyclability. Despite the strong interest in the field, the mechanisms of Li plating and stripping at the anode are not well observed and not yet fully understood [1]. The study of these phenomena would bring new insights to mechanical behavior during cycling and therefore may lead to optimization approaches at the cycle life level. In this purpose, we investigated the morphological change of SSM during the charge/discharge cycle. We looked at the influence of the cathode thickness and the packaging. Thick LiCoO_2 layers were used for the study with thickness from 5 to 30 μm . Volume variations upon cycling were followed thanks to a Neox microscope 3D (Sensofar) and were correlated to charge/discharge capacity.

Microbatteries are consisting of LiCoO_2 (LCO) cathode and LiPON electrolyte. The anode-free configuration was adopted for integration compatibility. The anode is formed in-situ during the first charge operation. Wafers with different LCO thickness and back end level have been carried out. Confocal microscope was setup in Ar-filled glovebox to monitor the volume expansion during the charge and discharge operations. SSM cycling was operated through Biologic instruments. Figure 1 depicts a schematic representation of the SSM and the setup. Images were acquired during chrono-amperometric full charge at 4.2 V and galvanostatic full discharge at $I_{\text{discharge}} = -3.5 \mu\text{A}$ within 3-4.2 V.

Charge/discharge operations and corresponding in-situ thickness variations are presented in Figure 2 for the samples with different LCO thickness. The complete charge is achieved when current reaches a threshold of 0.5 μA . Charge capacity linearly increased with cathode thickness: $Q_{\text{charge}} = 2.0 \mu\text{A.h}$ for the 5- μm -LCO and $Q_{\text{charge}} = 9.6 \mu\text{A.h}$ for the 30- μm -LCO. Thickness variation increase follows the same evolution as capacity during charge and discharge. Regardless of the cathode thickness, same plating and stripping speed are observed during charge and discharge operation, respectively. According to the Q_{charge} and the volume variation, a theoretical lithium density at the anode side has been observed for all LCO thickness.

Figure 3 depicts 2D images of 5- μm -LCO SSM at the onset of the charge. Pristine SSM presents a smooth surface (Fig. 3.a). Li plating is initiated with nucleated grains

(Fig. 3.b). Then, grains grow and tend to coalesce with each other, forming a uniform and smooth Li layer (Fig. 3.c). Figure 4 reports 2D images of 30- μm -LCO SSM at different states of cycling. Bumps at the edge formed during the charge are due to SSM device architecture: electric field bends and the increased ion flux induces a more important Li accumulation on the edge. Otherwise, a uniform and fully dense Li metal deposition is realized in solid-state configuration under zero external pressure [2]. At the end of discharge, the battery thickness is higher than the pristine one (+ 3.7 μm). The Li plating non-reversibility phenomena after the first charge is due to low diffusion coefficient at low state-of-charge [3].

Figure 5 shows the electrical behavior (Fig 5.a) and the volume expansion (Fig 5.b) of a SSM over 10 cycles. After the first cycle, cycling is performed at $\approx 100\%$ coulombic efficiency, demonstrating high reversibility of the phenomena and so high cyclability of SSM [3].

Finally, we followed the influence of the encapsulation layers on the volume variation. 2D images of the SSM reveals same volume variation for both samples. Those observations suggest that the encapsulation layers has no significant mechanical effect on the Li plating.

In-situ confocal microscopy under Ar atmosphere has been proved efficient to study the volume expansion of SSM upon cycling and is a promising technique to investigate failure mechanisms of SSM.

[1] J. Park *et al.*, « In situ atomic force microscopy studies on lithium (de) intercalation-induced morphology changes in Li_xCoO_2 micro-machined thin fi lm electrodes q », *J. Power Sources*, vol. 222, p. 417–425, 2013, doi: 10.1016/j.jpowsour.2012.09.017.

[2] D. Cheng *et al.*, « Freestanding LiPON: from Fundamental Study to Uniformly Dense Li Metal Deposition Under Zero External Pressure », *arXiv.org*, 8 août 2022. <https://arxiv.org/abs/2208.04402v2>.

[3] J. Celè, S. Franger, Y. Lamy, et S. Oukassi, « Minimal Architecture Lithium Batteries: Toward High Energy Density Storage Solutions », *Small Weinh. Bergstr.*

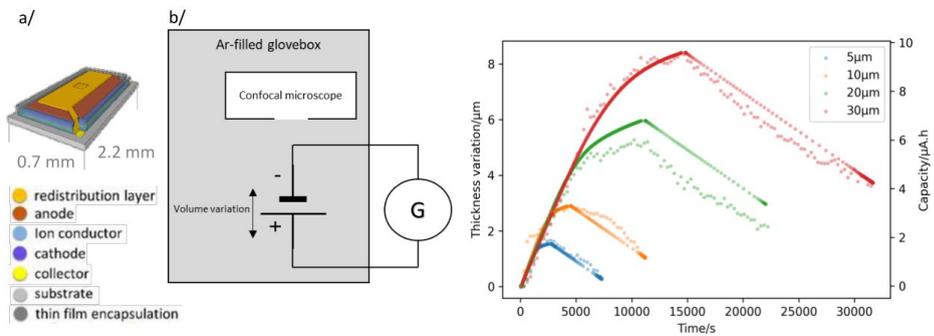


Figure 1: Schematic representations of a) a SSM and b) the setup

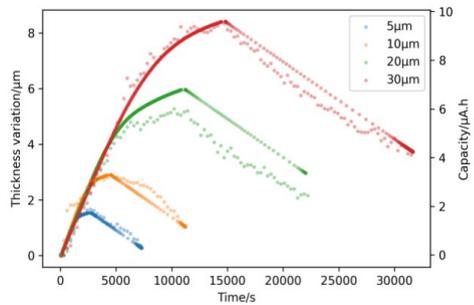


Figure 2: Capacity during charge/discharge operations (lines) and the correspond thickness variation (dots) for SSM with different LCO-thickness

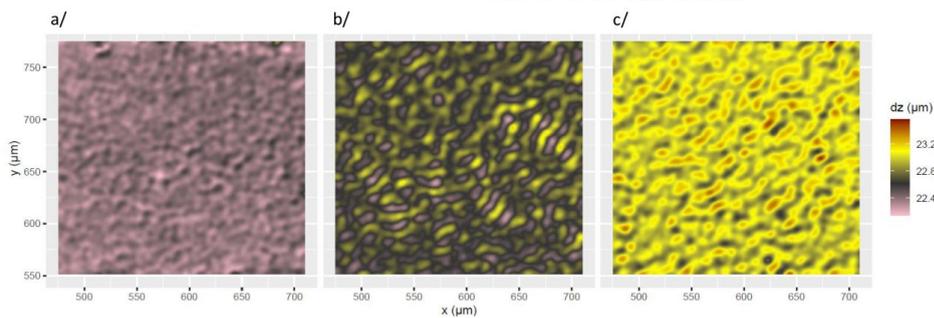


Figure 3: The first three 3D images of the surface of SSM with 5- μm -LCO cathode at the onset of the first charge operation.

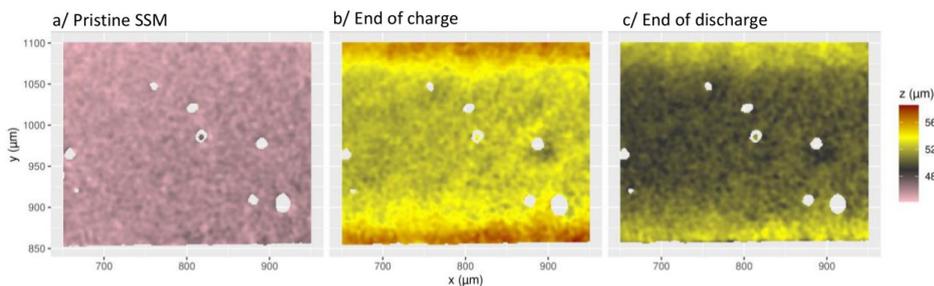


Figure 4: 3D images of the SSM part with 30- μm -LCO and no back end at different stage of charge/discharge: a/ before the first charge (pristine SSM), b/ at the end of charge, c/ at the end of discharge. LCO particles have been removed for the images processing.

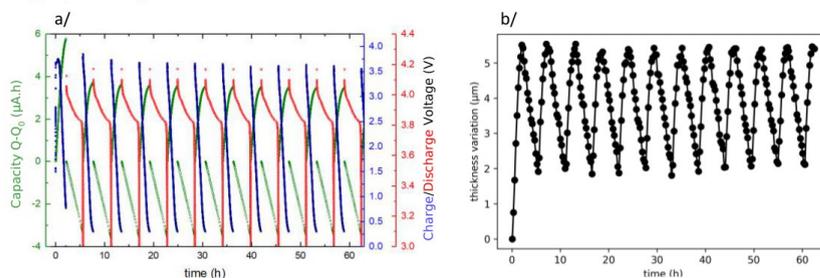


Figure 5: a/ Charge (red lines), discharge (blue lines) and corresponding capacities (green lines) of 20- μm -LCO SSM with back end b/ corresponding thickness variations. High reversibility of Li plating and stripping of SSM is observed.

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