

## Laayoune lithium-iron-phosphate batteries lfp

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Schematic illustration of uniformly distributed cathode components (left) and all solid-state lithium metal battery SS-LMB (right).

In order to understand the electrochemical behaviour of cathode, the other components in a battery such as electrolyte (SPNE) and lithium metal anode are used as a reference standard throughout the measurements. The EO/Li ratio of PEO/LiTFSI is maintained as 20:1 according to the literature report<sup>22</sup>, due to its good mechanical stability, high lithium ion transport number and high ionic conductivity. To start with, the cathode composition was adapted from the reported literature, wherein Judez et al.<sup>22</sup> have used 63 wt% of LFP as active material. Thus, the first LFP cathode (LFP-1) has a wt% composition of LFP:CB:PEO:LiTFSI = 63:7:22.7:7.3, where CB stands for super C65 conductive carbon black (see Table 1).

(a) and (b) are the SEM images, (c) and (d) are the cross-sectional SEM images of LFP-1 cathode before and after calendaring process, (e) comparative rate capability curve and (f) the corresponding charge/discharge curves of Li/SPNE/LFP cell using LFP-1 measured at 70 °C at different current rates, from 0.1C to 1C.

(a) Comparative rate capability curves of Li/SPNE/LFP cells having LFP-1 to LFP-3 cathodes and (b) the corresponding charge/discharge curves for LFP-3 cathode measured at 70 °C at different current rates.

(a) and (b) are the SEM images of calendered LFP-3 and LFP-4 cathodes prepared by using magnetic stirring and ball milling processes, (c) and (d) are the SEM back scattered images, (e) and (f) are the particle size distribution with color codes defining the sizes and (g) and (h) are the corresponding projected area analysis of LFP-3 and LFP-4 cathodes indicating the number of particles.

(a) Comparative cycle performance curves of Li/SPNE/LFP batteries having LFP-3 and LFP-4 as cathodes measured at 70 °C at 1C rate with inset of their corresponding charge-discharge curves for the 1st and 50th cycles, (b) the corresponding Nyquist plots before cycling and (c) a comparison of the eDRT time constants curves of the batteries (top) with symmetrical cathode-only cell (middle) and anode-only cells (bottom), all measured at 70 °C.

Further, we also studied the influence of different electrically conductive additives (super C65 conductive carbon black, CB and conductive graphite, CG) and their weight ratios on the electrochemical performance of

LFP cathode for SS-LMBs. It is reported in the literature for liquid based LIB system that the addition of graphite to LFP cathode helps in increasing the density of the electrode in comparison with super P carbon alone<sup>8</sup>. In addition, the large graphite particle size provides better contact between the active LFP particles and increases the electron percolation pathways more efficiently during fast reaction kinetics. Therefore, in the present study we examined the influence of homogenously distributed graphite particles in LFP cathode in the following.

The total amount of super C65 conductive carbon black (CB) used during the preparation of LFP1-4 cathode is 7 wt%. Therefore, the CB content is decreased systematically and the reduced amount is compensated by conductive graphite (CG) in the series, LFP5-7 and fully replaced by CG in LFP-8. The corresponding electrode compositions with different conductive additives are given in the Table 3.

(a) and (b) are the SEM and cross-sectional image of LFP-5 cathode (c) comparative cycle performance of Li/SPNE/LFP cells using LFP-4 to LFP-8 cathodes measured at 1C rate at 70 °C, (d) and (e) are the comparative graph of specific capacity vs. weight of conductive additives in the electrodes.

(a) Cycle performance curves of Li/SPNE/LFP cells using LFP-5, LFP-9 and LFP-10 cathodes measured at 1C rate at 70 °C, (b) the corresponding first charge/discharge curves, and (c) comparative specific capacity vs different weight ratios of LFP content in the electrode.

(a) Long-term cycle performance curve of Li/SPNE/LFP-5 cell with different thickness of SPNE (40, 90 and 130  $\mu$ m) measured at 2C rate at 70 °C and (b) Cycle performance curve of the same cell measured at 0.2C rate at 30 °C with in-set of its charge/discharge curves for 40  $\mu$ m thickness of SPNE.

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