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Processing and development of quasi-anode free lithium-based batteries by thermal evaporation

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The increasing popularity of portable electronic devices and the fast development of electric vehicles have greatly spurred the modernization of lithium-ion batteries (LIBs) towards higher energy density, higher power density, and better safety. The currently commercialized graphite anode is limited by its relatively low theoretical and practical capacity (372 mAh g⁻¹), its sluggish lithium-ion diffusivity and the difficulties to meet the growing demand for the next-generation energy storage systems.[1] As superior alternatives to graphite anode, lithium metal anodes display higher theoretical capacities. However, these systems produce dendrite and whisker formations. These, together with volumetric changes, strongly affect the safety and lifetime of the battery.[2]

In this sense, the development of anode-less batteries provides numerous benefits, which can be highlighted as an increased energy density, safety improvement and reduction of costs. Many of the current batteries under the concept of anodes-less or anode free, have an extra reservoir of lithium to compensate the irreversibility of the electrochemical processes. This extra contribution is usually given by the cathode with the use of sacrificial salts; in this case, preconditioning cycles are needed with the subsequent increase in fabrication costs and reduce the energy density. On the other hand, surface modification of the current collector (CC) is one of the most used strategies to improve electrochemical behavior in anode free lithium metal batteries (AFLMB).[3] In this regard, thermal evaporation is a well-known and very basic technique for coating surfaces and substrates with both organic and inorganic thin layers.[4] This technique is especially suitable for materials with low melting points and high vapor pressure, as for instance lithium with melting point 180°C.[5]

In this study, a quasi-anode free lithium metal battery is presented. It has been fabricated by using thermal evaporation technique to deposit a lithium thin film onto the Cu current collector. No modifications have been carried out neither any CC treatment. Cell assembly has been done with commercial liquid electrolyte without additives.

This study will show the substantial improvements on the electrochemical performance of lithium batteries when used with lithium thin film deposited on Cu current collector by thermal evaporation.

[1] D. Lin, Y. Liu, and Y. Cui, "Reviving the lithium metal anode for high-energy batteries," *Nat Nanotechnol*, vol. 12, no. 3, pp. 194–206, Mar. 2017, doi: 10.1038/nnano.2017.16.

[2] H. Wang, J. Wu, L. Yuan, Z. Li, and Y. Huang, "Stable Lithium Metal Anode Enabled by 3D Soft Host," *ACS Appl Mater Interfaces*, vol. 12, no. 25, pp. 28337–28344, Jun. 2020, doi: 10.1021/acsami.0c08029.

[3] C.-J. Huang et al., "Lithium Oxalate as a Lifespan Extender for Anode-Free Lithium Metal Batteries," *ACS Appl Mater Interfaces*, vol. 14, no. 23, pp. 26724–26732, Jun. 2022, doi: 10.1021/acsami.2c04693.

[4] T. Nishinaga, "Handbook of Crystal Growth: Thin Films and Epitaxy: Second Edition," *Handbook of Crystal Growth: Thin Films and Epitaxy: Second Edition*, vol. 3, pp. 1–1346, Dec. 2014, doi: 10.1016/C2013-0-09792-7.

[5] B. Acebedo et al., "Current Status and Future Perspective on Lithium Metal Anode Production Methods," *Adv Energy Mater*, vol. 13, no. 13, Apr. 2023, doi: 10.1002/aenm.202203744.

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