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Real-Time Interfacial Analysis in Li-ion Batteries via Operando SHINERS

The development of new electrode materials for lithium or sodium-ion batteries, is usually hampered by the electrode/electrolyte interfacial instability upon cycling, which leads ineluctably to performance degradation. The repeated formation and dissolution of electrode passive layer upon cycling, known as the **SEI/CEI** layer, strongly contributes to the battery capacity fading by consuming cyclable lithium. The understanding of the SEI/CEI layer properties is key to optimize both the electrode and electrolyte formulations toward improved performances. However, only a handful of spectroscopy techniques have the sufficient sensitivity, resolution (temporal, spatial, chemical) to characterize very thin evolving interfacial layers (10s of nm) buried in the battery assembly. If Raman has been largely applied to the characterization of battery electrode materials, it is totally blind to the SEI/CEI interfacial layer developed in LIBs.

This work focusses on the development of enhanced Raman spectroscopy (Shell Isolated Nanoparticle Enhanced Raman Spectroscopy: **SHINERS** [1]) to capture the dynamics of formation and composition of the SEI/CEI layer on promising electrode materials (Sn, Si) during battery operation (operando). Toward this aim, specific strategies have been developed in our group to access the interface buried in the battery assembly and also to enhance the interfacial signal while minimizing that of the electrode material and of the electrolyte ("bulk" Raman and autofluorescence signals).

References

[1] Li, J. F., Tian, X. D., Li, S. B., Anema, J. R., Yang, Z. L., Ding, Y., Wu, Y. F., Zeng, Y. M., Chen, Q. Z., Ren, B., Wang, Z. L., & Tian, Z. Q. Nature Protocols, 8(1), (2013), 52–65.
[2] Gajan, A., Lecourt, C., Torres Bautista, B. E., Fillaud, L., Demeaux, J., & Lucas, I. T, ACS Energy Lett, 44 (2021) 994–998.

Figures

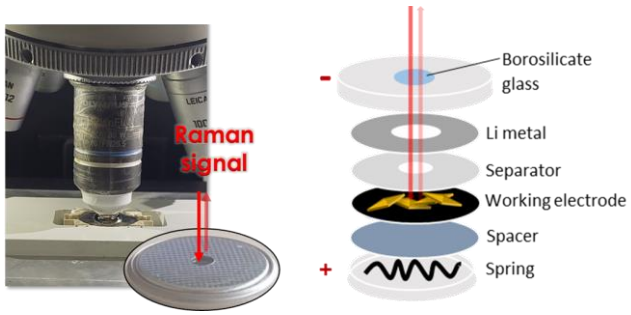

Figure 1: Typical schematic depiction of the spectro-electrochemical coin-cell design for the operando SHINERS characterization of the electrode/electrolyte interfacial processes taking place in operating Li-ion batteries.