In-situ characterization of NMC particles with applications in Li-ion batteries

Javier García-Alonso[1]
S. Nahirniak,[2] D. Maestre,[2] B. Saruhan,[2] A. Cremades,[1] B. Méndez[1]
[1] *Dpto. Física de Materiales, Facultad de CC. Físicas, Universidad Complutense de Madrid, 28040, Spain*
[2] *Institute of Materials Research, German Aerospace Center (DLR), Linder Hoehe 51147, Cologne, Germany*
jgarci13@ucm.es

The advent of modern Lithium-ion batteries (LiBs) and energy storage technologies will rely on innovative materials and improved characterization techniques in the search of high-energy and long-term cycling LiB components [1, 2]. Amongst the most promising materials used as cathodes, Ni-Mn-Co (NMC)-based compounds with core-shell structure are gaining increased attention owing to their high specific capacity and thermal stability. The characterization of these materials requires of advanced microscopy and spectroscopy techniques, including *in-situ* and *in-operando* techniques, in order to achieve deeper comprehension of the phenomena governing the battery performance.
In this work, NMC core-shell microparticles have been studied by a combination of techniques including scanning electron microscopy (SEM), X-ray diffraction (XRD), thermo-XRD, energy dispersive x-ray spectroscopy (EDS), Raman spectroscopy, and X-Ray Photoelectron Spectroscopy (XPS), including i*n-situ* characterization as well. The combination of these techniques allows to achieve insights in the morphological, structural, compositional, and electronic properties of the NMC microparticles under study. The analysed microparticles exhibited a rounded appearance with an average diameter of 4 µm and variations in their morphology and properties as a function of the synthesis route, as observed by SEM and XRD. The presence of Ni-rich core and a Mn-rich shell with submicrometric dimensions have been confirmed by EDS, while the formation of oxides presenting the usual layered structure of NMC (R-3m) together with the appearance of spinel phases as a function of the synthesis procedure have been assessed by means of XRD and Raman spectroscopy. This layered structure is directly related to the high capacity and structural stability of NMC cathodes when the battery is cycled. Further characterization, including in-situ-thermo XRD, in -situ XPS under variable pressure, as well as in-situ SEM with variable temperature up to 900 ºC have been also carried out. These measurements confirm the formation of NiO as the temperature increases up to 900 ºC, as well as the promotion of changes in the morphology of the microparticles. Finally, in-situ XPS measurements at temperatures up to 500 ºC and a variable pressure, indicate variations in the Mn(3s) and O(1s) core levels, as well as in the electrical conductivity of the samples. The combination of these results, including the in-situ characterization, can lead to achieve a deeper knowledge of the properties of the NMC microparticles in the search of improved performance in LiBs.

References

[1] J. Amici, P. Asinari, E. Ayerbe et al. *Adv. Energy Mat*. 2101785 (2022) "A roadmap for transforming research to invent the batteries of the future designed within the European Large scale research initiative Battery 2030+".
[2] A. Jena, Z. Tong, Z. Bazri, K. Iputera et al. *J. Phys. Chem. C*, 125, 16924 (2021) "In situ/operando methods of characterizing all—solid-state Li ion batteries: understanding Li-ion transport during cycling".