

Supporting Information

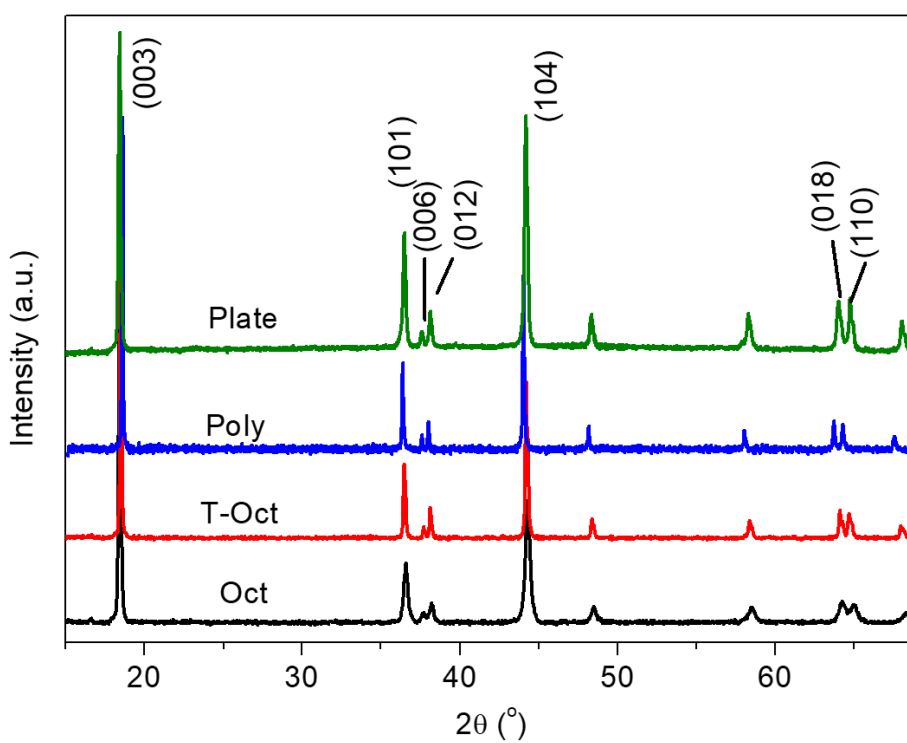


Figure S1. XRD patterns of a) Oct, b) T-Oct, c) Poly and d) Plate crystal samples.

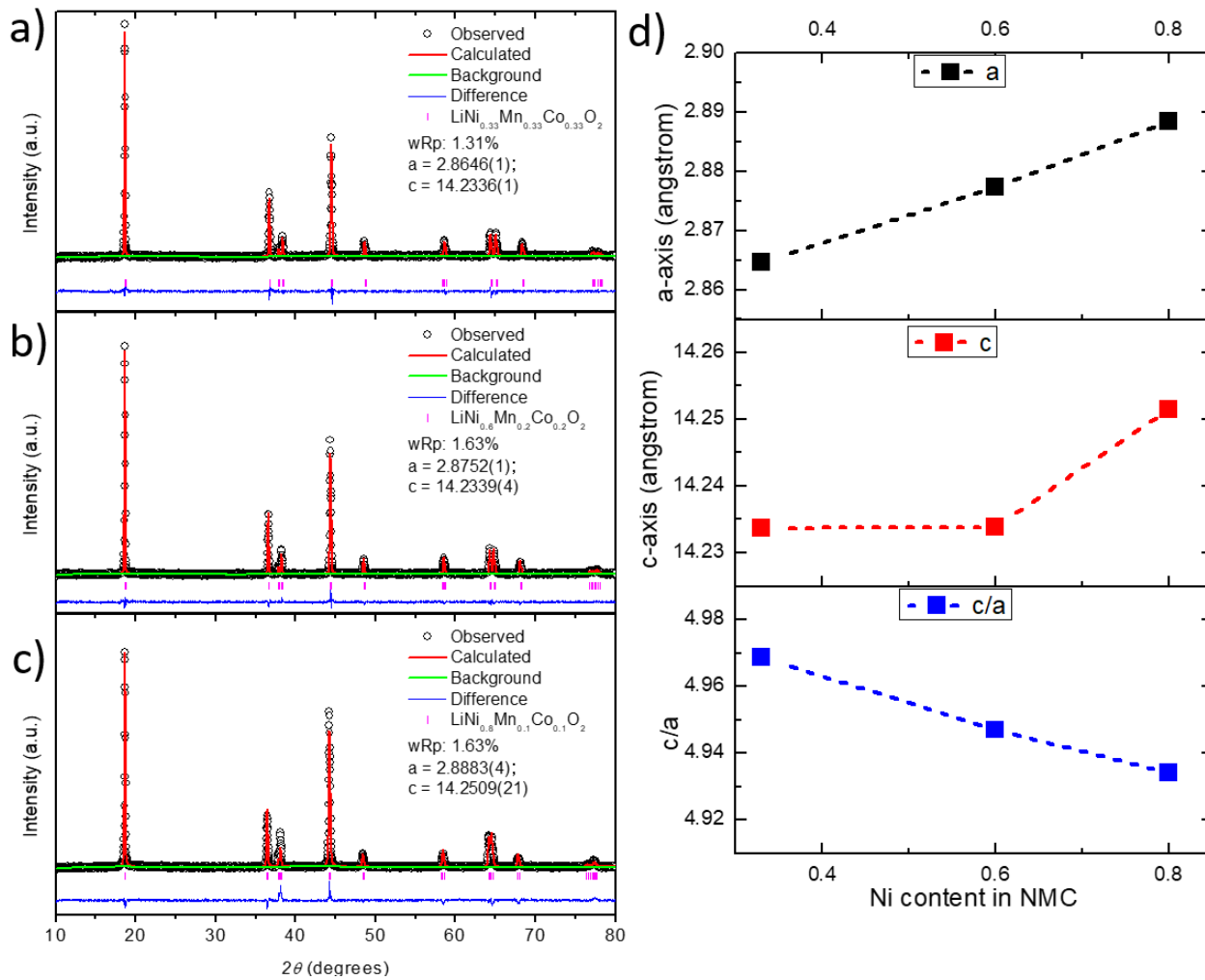


Figure S2. a-c) Rietveld refinement of XRD patterns collected on 1 μm T-Oct NMC333, NMC622 and NMC811 and d) changes in lattice parameters a , c and c/a ratio as a function of Ni content in the NMCs.

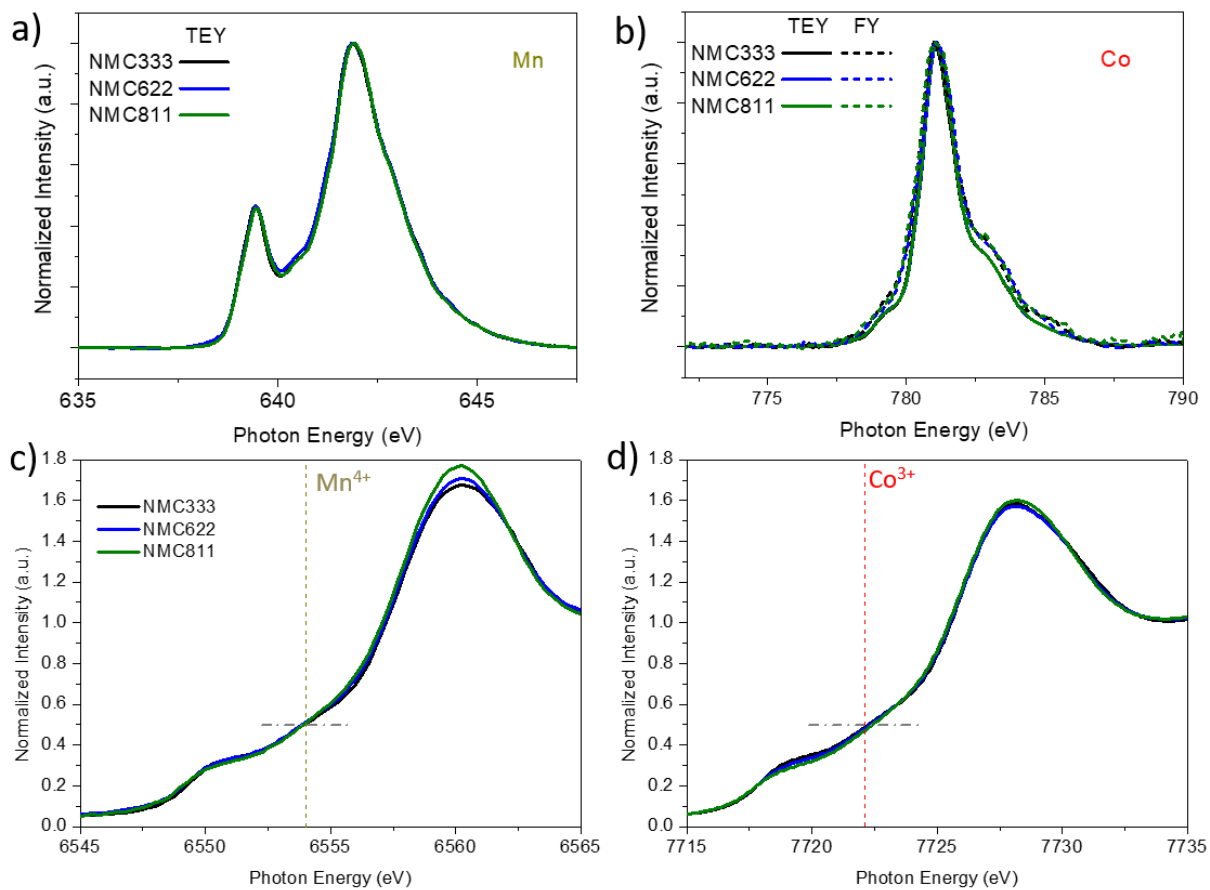


Figure S3. a-b) Mn and Co L_3 -edge sXAS spectra and c-d) K -edge hard XAS spectra collected on 1 μm T-Oct NMC333, NMC622 and NMC811 samples.

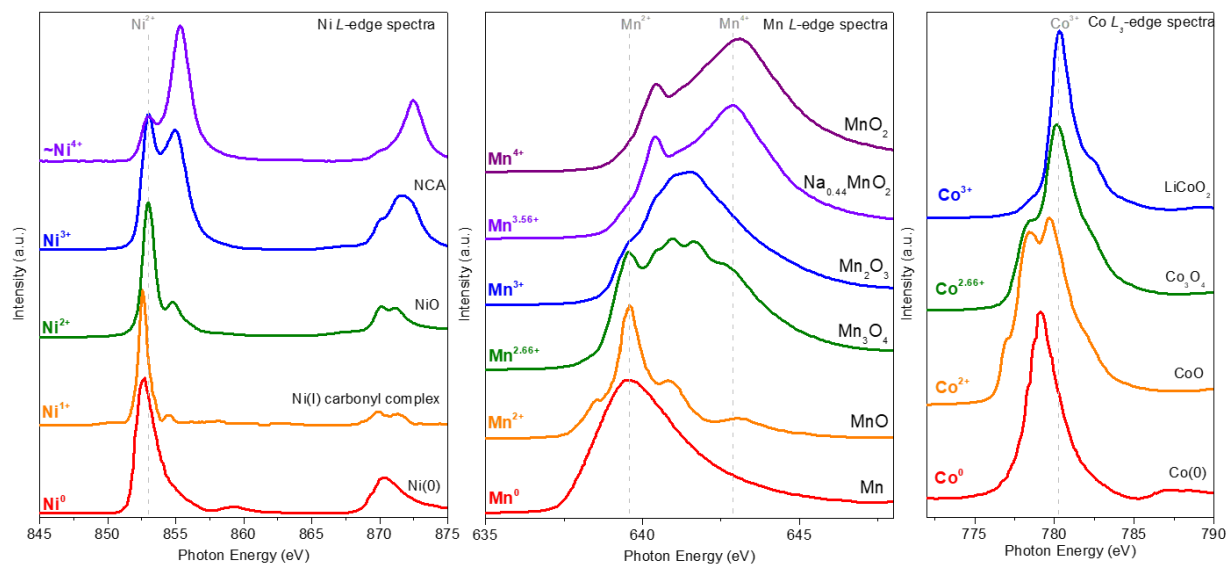


Figure S4. Soft XAS *L*-edge reference spectra of Ni, Mn and Co at different oxidation states.

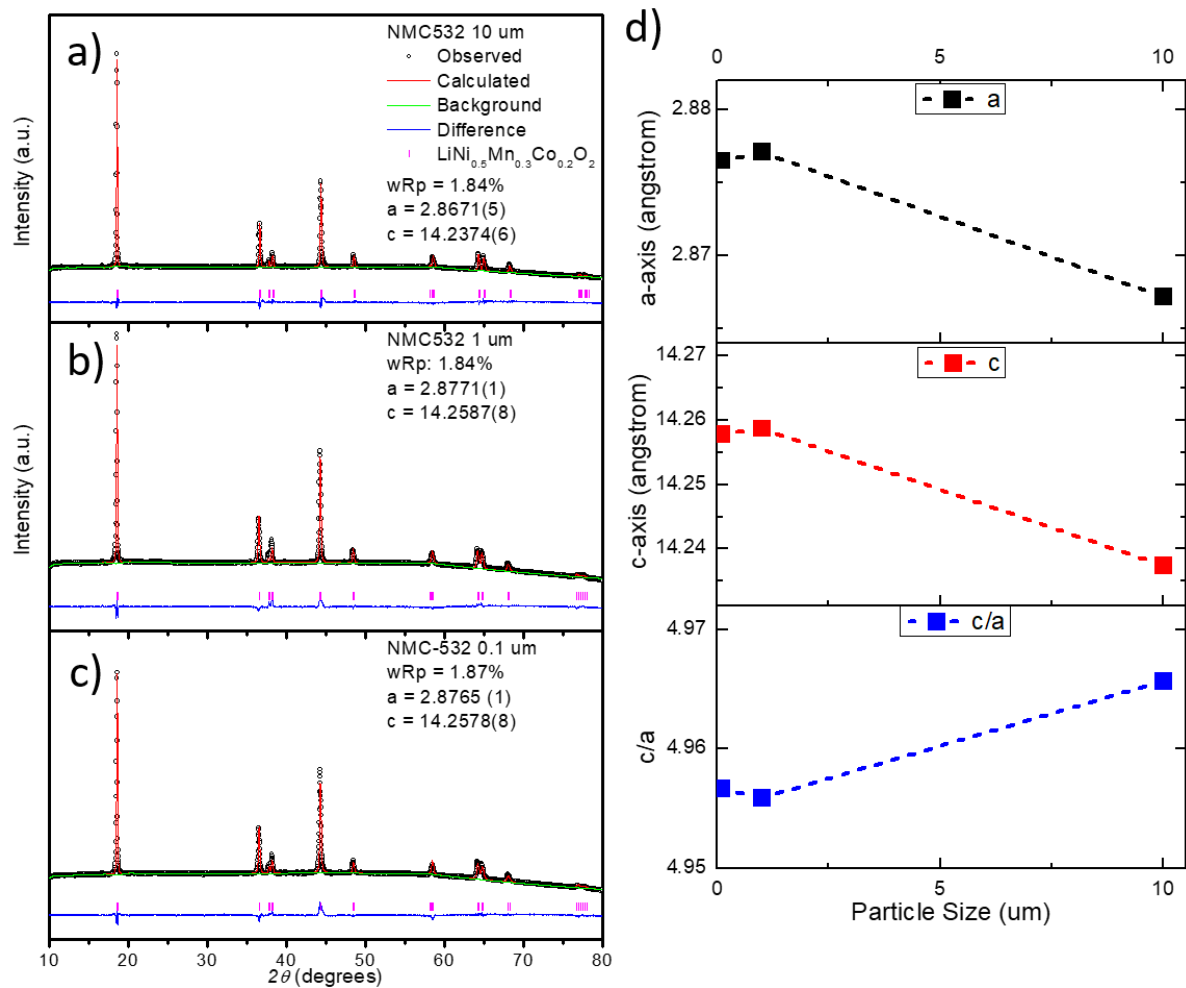


Figure S5. Rietveld refinement of XRD patterns collected on T-Oct NMC532 crystal samples with a size of: a) 10 μm, b) 1 μm, c) 0.1 μm, and d) changes in lattice parameter a , c and c/a as a function of particle size.

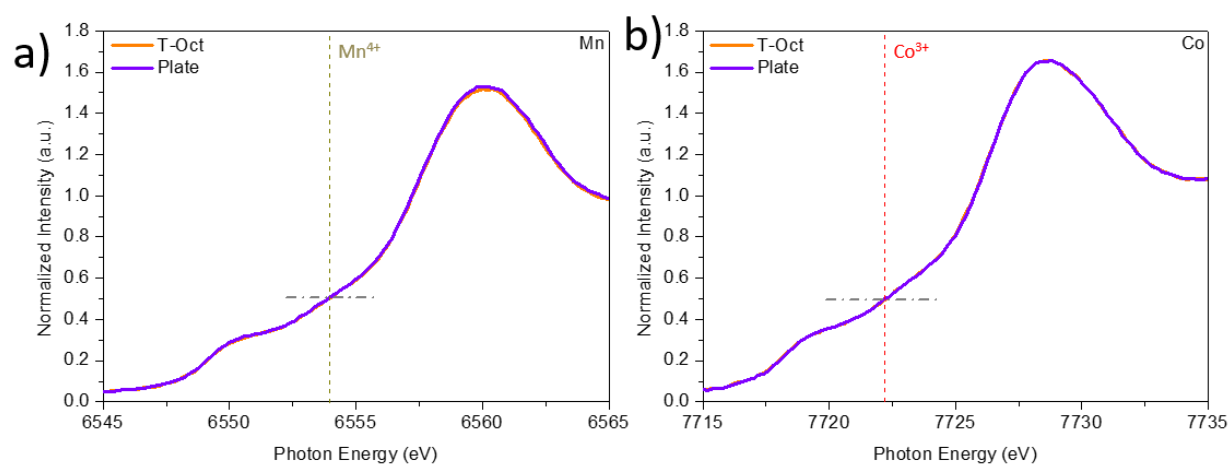


Figure S6. a) Mn and b) Co hard XAS *K*-edge spectra of Plate and T-Oct NMC333 crystal samples, respectively.

Table S1. Chemical composition of NMC samples determined by ICP.

Samples			Chemical composition
Composition	Morphology	Size (μm)	
333/622	Oct	10	$\text{Li}_{0.97}\text{Ni}_{0.34}\text{Mn}_{0.33}\text{Co}_{0.33}\text{O}_2 / \text{Li}_{0.94}\text{Ni}_{0.60}\text{Mn}_{0.19}\text{Co}_{0.21}\text{O}_2$
532	T-Oct	10	$\text{Li}_{1.02}\text{Ni}_{0.51}\text{Mn}_{0.30}\text{Co}_{0.21}\text{O}_2$
333/622	T-Oct	1	$\text{Li}_{0.95}\text{Ni}_{0.35}\text{Mn}_{0.32}\text{Co}_{0.33}\text{O}_2 / \text{Li}_{0.95}\text{Ni}_{0.61}\text{Mn}_{0.20}\text{Co}_{0.19}\text{O}_2$
532	T-Oct	1	$\text{Li}_{0.97}\text{Ni}_{0.51}\text{Mn}_{0.30}\text{Co}_{0.21}\text{O}_2$
532	Poly	1	$\text{Li}_{0.99}\text{Ni}_{0.50}\text{Mn}_{0.31}\text{Co}_{0.21}\text{O}_2$
532	T-Oct	0.5	$\text{Li}_{0.94}\text{Ni}_{0.51}\text{Mn}_{0.32}\text{Co}_{0.19}\text{O}_2$
333/532/622	T-Oct	0.1	$\text{Li}_{1.03}\text{Ni}_{0.34}\text{Mn}_{0.33}\text{Co}_{0.31}\text{O}_2 / \text{Li}_{0.97}\text{Ni}_{0.51}\text{Mn}_{0.30}\text{Co}_{0.21}\text{O}_2 / \text{Li}_{0.93}\text{Ni}_{0.58}\text{Mn}_{0.21}\text{Co}_{0.20}\text{O}_2$
333	Plate	0.1	$\text{Li}_{1.00}\text{Ni}_{0.32}\text{Mn}_{0.34}\text{Co}_{0.34}\text{O}_2$
622	Plate	0.1	$\text{Li}_{0.98}\text{Ni}_{0.63}\text{Mn}_{0.19}\text{Co}_{0.18}\text{O}_2$