Transformations in Li-rich layered oxides have been extensively studied recently for their potential application in Li-ion batteries. These materials have attracted a lot of interest due to the high capacity offered by them. However, the structure of these materials in their pristine state is not clearly understood. Several reports have assigned their structure to be trigonal (R-3m), monoclinic (c2/m), or a combination of both (composite). The present study discusses the structure of \( \text{Li}_{1.2} \left( \text{Ni}_{0.13} \text{Mn}_{0.54} \text{Co}_{0.13} \right) \text{O}_2 \) prepared with two different morphologies: plates and needles, using the results obtained from aberration corrected (scanning) transmission electron microscopy, electron energy loss spectroscopy (EELS), convergent beam electron diffraction and precession electron diffraction tomography and question the validity of the the claims of them being “composite”. It was found that these materials consist of domains which correspond to variants of monoclinic structure. It will be shown how diffraction-based experiments on such materials can often lead to misleading conclusions, since analysis of diffraction-based techniques inevitably assign them as trigonal, although the present study shows that the three-fold symmetry observed in electron diffraction patterns result from the combination of the variants having monoclinic structure.

Furthermore, results from STEM and EELS experiments showed that the pristine materials have several defects. The plates exhibited a differently ordered structure on their surface, and the needles exhibited several cobalt-rich line defects. These results prescribe that extreme care should be taken while interpreting the electron microscopy results obtained from cycled samples.

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Fig. 1: STEM HAADF image showing the structure of monoclinic Li$_{1.3}$(Ni$_{0.13}$Mn$_{0.54}$Co$_{0.13}$) O$_2$.

Fig. 2: Inverted and color-coded HAADF STEM image showing the three variants (in projection) of the monoclinic structure.