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Supporting Information

Under Pressure: Offering Fundamental Insight into Structural Changes on Ball Milling Battery Materials

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SI Figure 1: Fitted central transition peaks of single pulse spectra for (a) BM-Li₂MoO₄ and (b) parent Li₂MoO₄. The fitting parameters obtained are summarised in table (c).

SI Table 1: Approximate time needed to achieve complete conversion of Li_2MoO_4 from phenakite to spinel phase by ball milling in the Pulverisette P23 ball mill under different milling conditions.

Frequency Ball size	50 Hz	40 Hz
10 mm	15 min	45 min
7 mm	45 min	120 min



SI Figure 2: Li_2MoO_4 cycled in the range of 3.0 – 0.5 V. Due to the programme safety limits, the cell reached 0.55 V rather than the desired 0.5 V.



SI Figure 3: PXRD data, Rietveld refinement (initial exposure) and calculated wt% of Sp-Li₂MoO₄ and Ph-Li₂MoO₄ (Cu Kα). The same sample was remeasured every 2 h to assess the rate of change. The start of the transition occurs within a couple of minutes of exposure to ambient conditions, before the rate slows considerably – most likely due to the initial phenakite shell formed limiting further transformation.

Note: the time increment refers to the amount of time that has passed at the start of the subsequent scan.

O ₂ mass calculation		
Starting mass (mg) 30.1		108
Maximum mass (%)	108.0	
Maximum mass (mg)	32.52	106
Mass difference (mg)	2.4210	(%) 101
Moles of Li ₂ MnO ₃	0.0002784	W Wass
Moles of O ₂	7.57E-05	102
Mole ratio	0.271	
O content in initial Li ₂ MnO ₃	2.46	100 -
Calc Mn oxi. state	2.9	98
		100 200 300 400 500 600 70 Temperature (°C)

SI Figure 4: TGA data and calculation to determine oxygen loss during the ball milling process of Li_2MnO_3 . Data suggests reduction of Mn^{4+} to Mn^{3+} has occurred during milling. The inset shows resultant colour change induced through milling.



SI Figure 5: TEM images of ball milled Li_2MnO_3 (a) and Li_2SnO_3 (b) particles. The centre images (b and e) shows the FFT on whole particle for BM- Li_2MnO_3 and BM- Li_2SnO_3 . HR-TEM (c and f) images reveals that there are localized crystalline structure in an amorphous matrix, where crystalline planes, though distorted, are clearly visible.



SI Figure 6: SEM images of pristine (a) and ball-milled (b) Li₂MnO₃. Images taken on the Zeiss EVO15 instrument.



SI Figure 7: SEM images of pristine (a) and ball-milled (b) Li₂SnO₃. Images taken on the ThermoFisher Scientific Apreo 2 SEM.

The electrochemical performance of Li_2MnO_3 is presented below in **SI Figure 8**. The electrochemical data of Li_2SnO_3 has not been included due to poor performance.



SI Figure 8: Voltage profile for BM-Li₂MnO₃ (8 h at 450rpm) between 1.5 and 4.8 V at 10 mA g⁻¹.



SI Figure 9: Resulting XRD patterns of $H-Nb_2O_5$ after ball milling at 50 Hz at set durations. Each measurement made use of a fresh sample (50 mg) and one 7 mm diameter ball. (Cu K α).



SI Figure 10: Resulting XRD patterns of H-Nb₂O₅ after ball milling at 40 Hz at set durations. Each measurement made use of a fresh sample (50 mg) and one 7 mm diameter ball. (Cu K α).



SI Figure 11: SEM images of H-Nb₂O₅ from ball milling at 50 Hz at set durations: a) pristine; 0 mins, b) 15 mins, c) 1h, d) 1h 30 mins, e) 2 h, and f) 3 h.



SI Figure 12: Resulting XRD patterns of $Li_{1.2}Ni_{0.4}Mg_{0.2}Mo_{0.2}O_2$ and DRS $Li_{1.2}Ni_{0.4}Mg_{0.2}Mo_{0.2}O_{1.8}F_{0.2}$, which shows a transformation from a cation ordered layered structure to a DRS structure on F doping. (Cu K α).