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## **Electronic Supplementary Information**

## Entropymetry for Non-destructive Structural Analysis of LiCoO<sub>2</sub> Cathodes

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**Fig. S1**. Correlation of  $\Delta S$  profile with voltage profile.



Fig. S2.  $\Delta S$  profiles of three pre-cycled Co100 cells during charging.



**Fig. S3**. Schematic diagrams of phase transitions of (a) LCO and (b) Ni-doped LCO during high voltage cycling.



Fig. S4.  $\Delta S$  profile of pre-cycled Co93 throughout charging.



Fig. S5 Open-circuit voltage changes upon temperature variation.

	Sample	Atomic ratio			
		Со	Ni		
	Co100	0.999	0.001		
	Co97	0.971	0.029		
	Co95	0.953	0.047		

**Table S1.** Elemental analysis of Co100, Co97 and Co95.

	Trough		Peak		Trough-to-peak height	
	Before cycles	After cycles	Before cycles	After cycles	Before cycles	After cycles
Co100	-75.79	-44.39	18.78	6.32	94.57	50.71
Co97	-49.45	-29.13	18.69	6.52	68.14	35.65
Co95	-28.42	-18.98	4.41	-2.88	32.82	16.10

**Table S2**. Calculation of trough-to-peak height change of Co100, Co97, and Co95 before and after cycling in the monoclinic phase region.