

## *Supporting Information*

# Design of Nickel-rich Layered Oxides Using *d* Electronic Donor for Redox Reactions

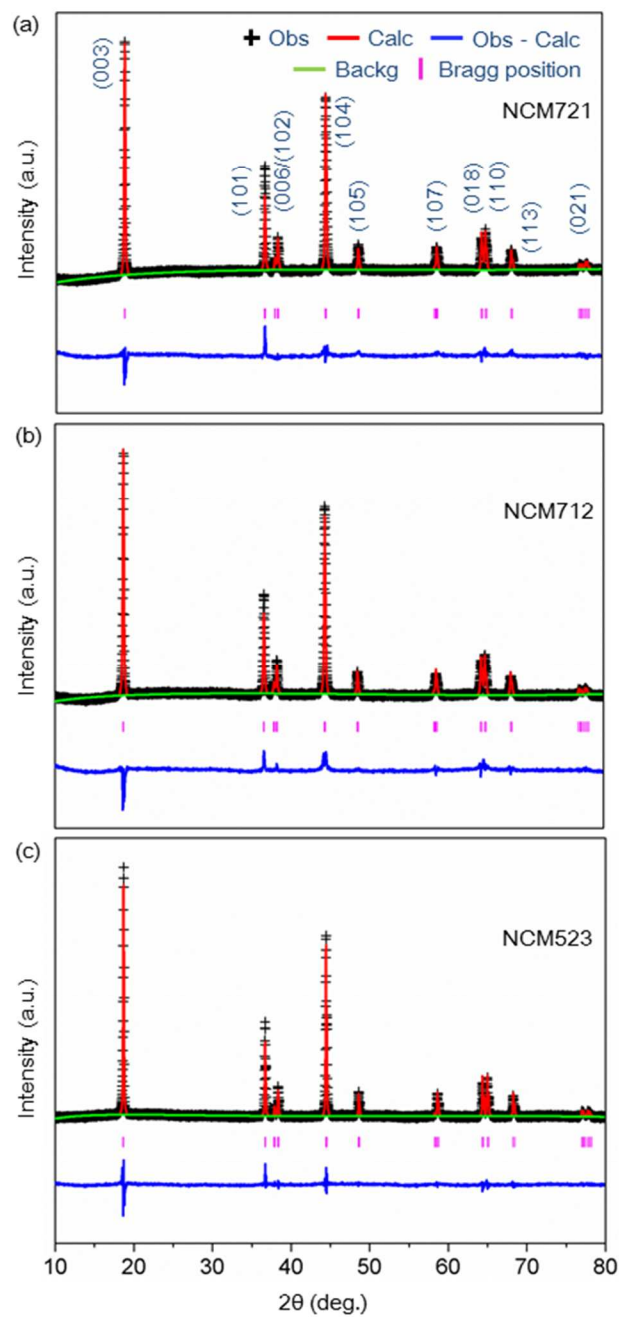
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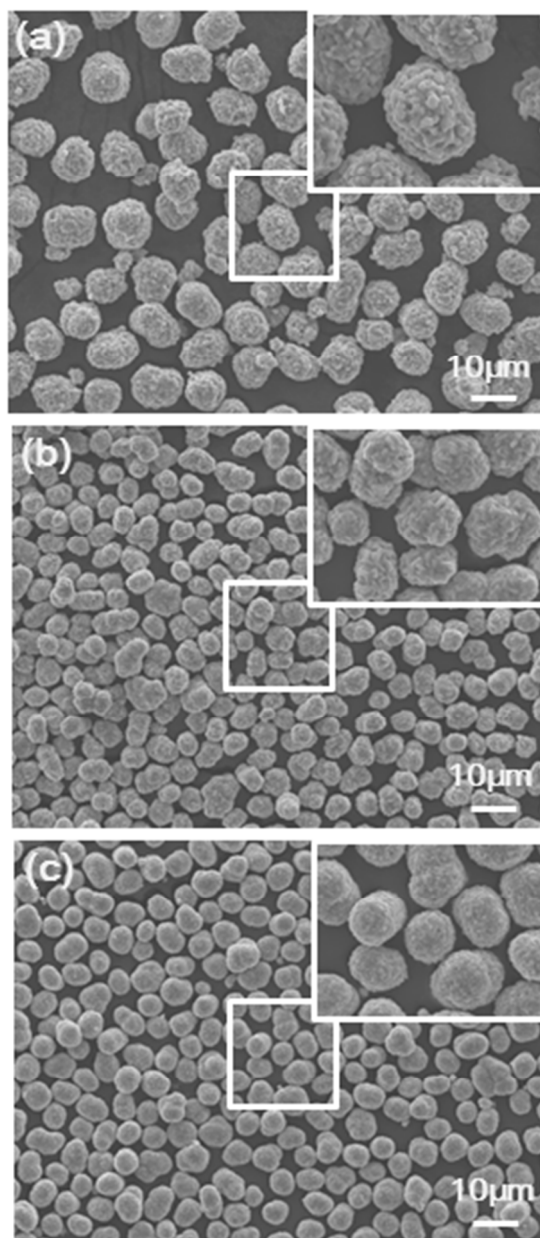
**Figure S1.** Rietveld refinement of X-ray diffraction (XRD) patterns of (a) NCM523, (b) NCM712, and (c) NCM721.

Composition	Lattice parameters				R <sub>wp</sub>	R <sub>exp</sub>
	a (Å)		c (Å)			
	Exp.	DFT	Exp.	DFT		
LiNi <sub>0.5</sub> Co <sub>0.2</sub> Mn <sub>0.3</sub> O <sub>2</sub> (NCM523)	2.8687	2.8740	14.2350	14.1172	2.61	1.25
LiNi <sub>0.7</sub> Co <sub>0.1</sub> Mn <sub>0.2</sub> O <sub>2</sub> (NCM712)	2.8738		14.2237		2.44	1.29
LiNi <sub>0.7</sub> Co <sub>0.2</sub> Mn <sub>0.1</sub> O <sub>2</sub> (NCM721)	2.8665	2.8664	14.1895	14.0666	2.69	1.17

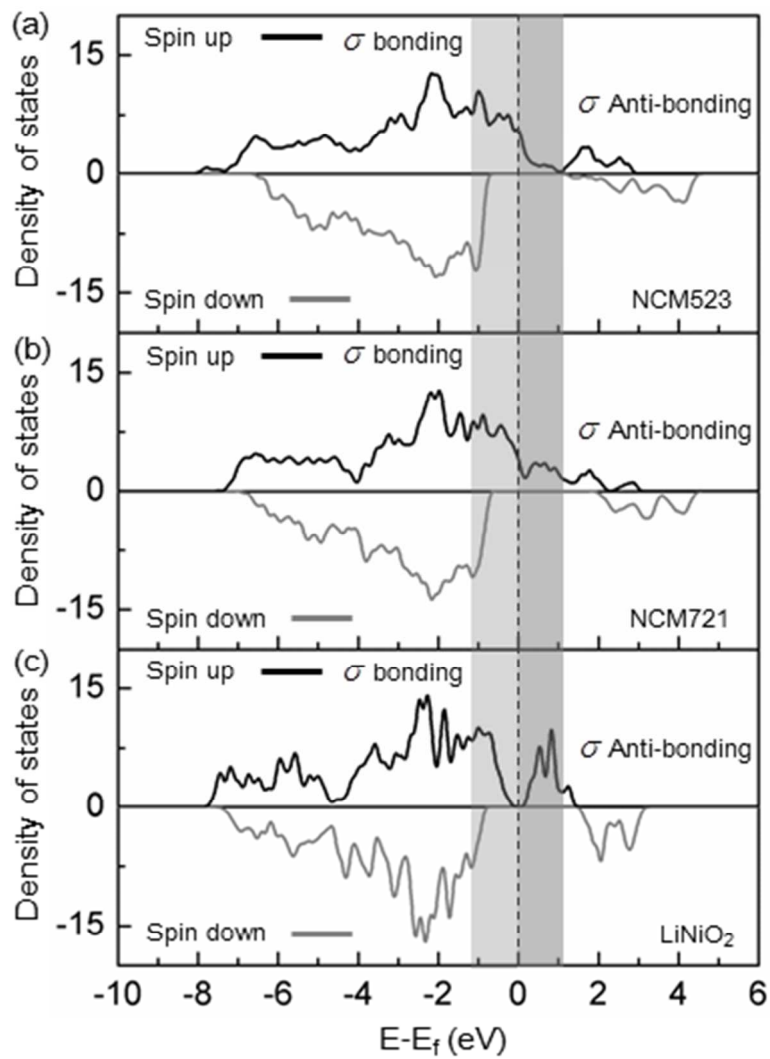
**Table S1.** Comparison of lattice parameters of the NCM layered structures obtained from experiment (Rietveld refinement results) and First-principles calculation.

Compounds	Elements	Content (ppm)	Weight (g)	Wt (%)	Mol (%)	Fraction
NCM523	Li	71182	13.92	7.12	200.50	1.09
	Ni	276213	54.00	27.62	92.01	0.50
	Co	113478	22.19	11.35	37.65	0.20
	Mn	154665	30.24	15.47	55.04	0.30
NCM712	Li	71441	13.97	7.14	201.23	1.04
	Ni	404976	79.17	40.50	134.90	0.70
	Co	60718	11.87	6.07	20.14	0.11
	Mn	106300	20.78	10.63	37.83	0.19
NCM721	Li	68686	13.43	6.87	193.46	1.04
	Ni	390132	76.27	39.01	129.95	0.70
	Co	108584	21.23	10.86	36.02	0.19
	Mn	55528	10.86	5.55	19.76	0.11

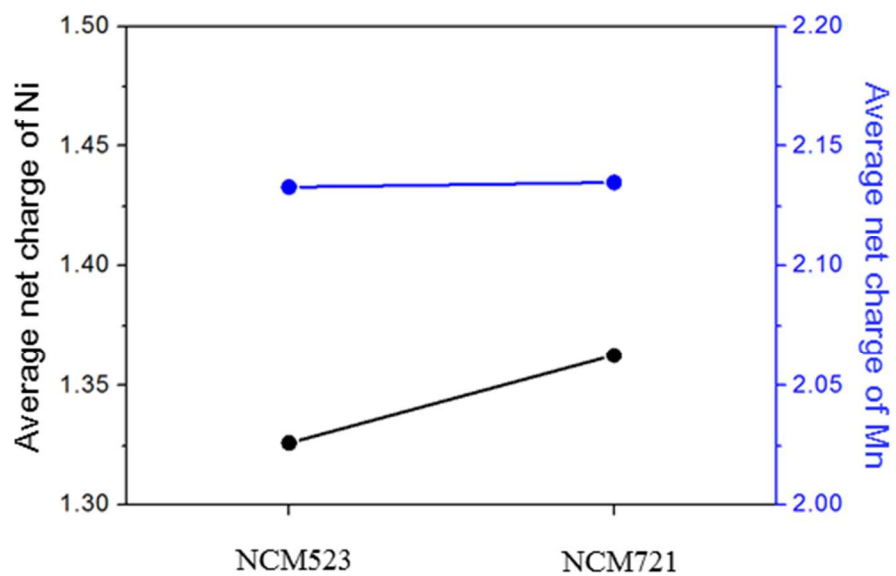
**Table S2.** Chemical compositions results of inductively coupled plasma mass spectroscopy (ICP-MS) for NCM523, 712 and 721.



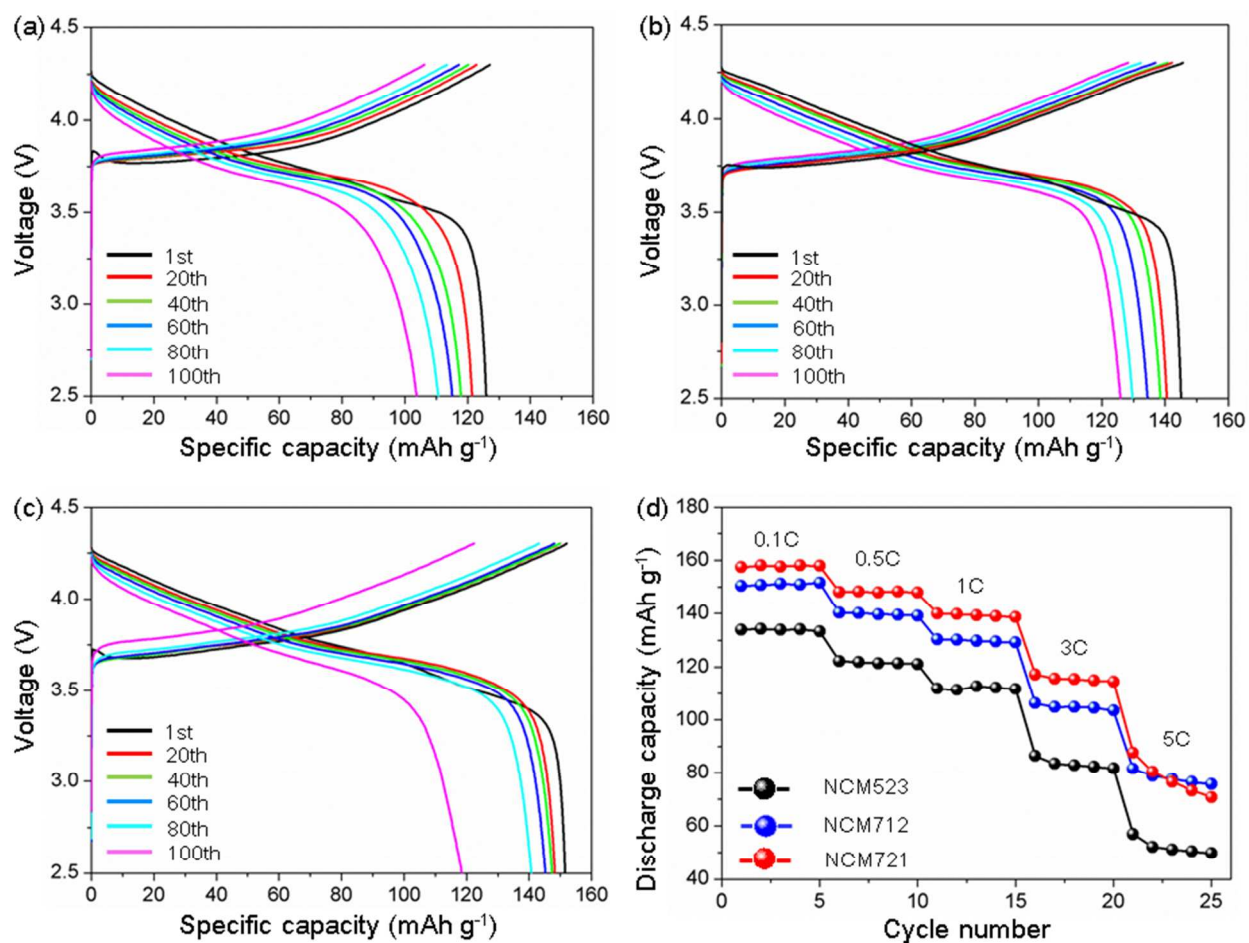
**Figure S2.** Field emission scanning electron microscope (FESEM) images of (a) NCM523, (b) NCM712, and (c) NCM721.



**Figure S3.** 2p-electron partial density of states (PDOS) of O ions in (a) NCM523, (b) NCM721, and (c) LiNiO<sub>2</sub>.



**Figure S4.** Average net charge of Ni ions (black circles) and Mn ions (blue circles) in NCM523 and NCM721.



**Figure S5.** Charge and discharge profiles of (a) NCM523, (b) NCM712, and (c) NCM721 with a constant specific current of 0.5 C rate in the voltage range of 2.5–4.3 V vs.  $\text{Li/Li}^+$ . (d) Rate performance of the three NCM samples.