Supporting Information

High-performance Li-ion Batteries using Nickel-rich Lithium Nickel Cobalt Aluminium Oxide-Nanocarbon Core-Shell Cathode: *In operando* X-ray diffraction

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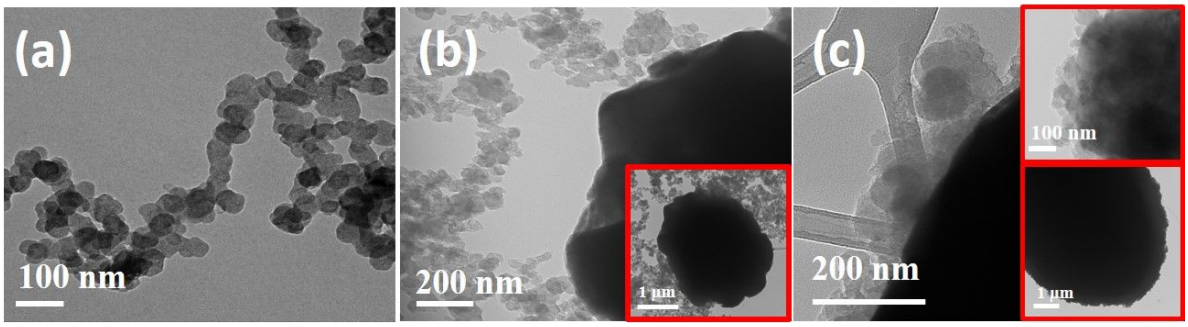

Figure. S1 HR-TEM images of (a) Conductive super P, (b) Loosely bounded super P on NCA obtained by physically blending (PNCA) and (c) Intact carbon coating by mechano-fusion (NCACS).

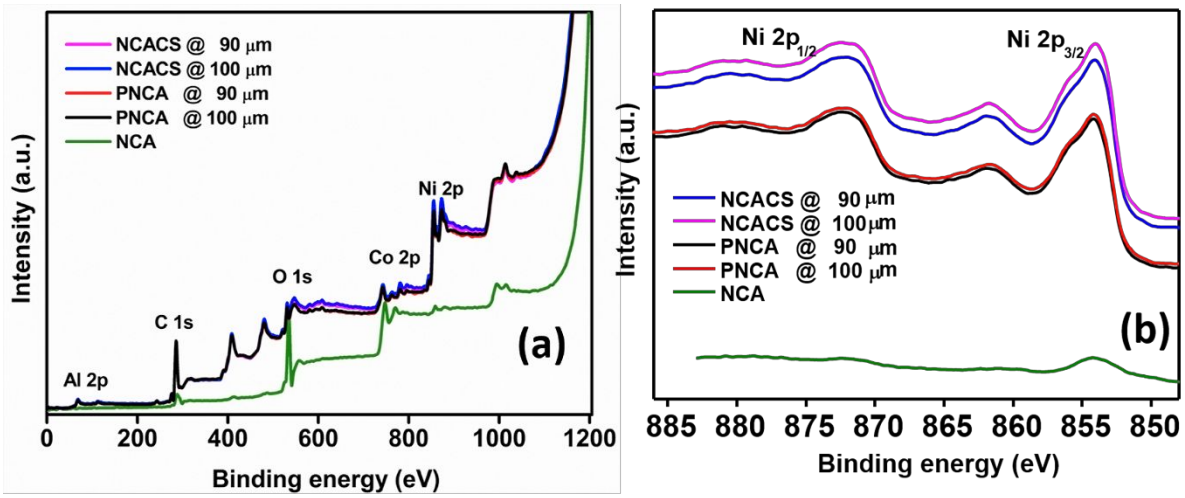

Figure. S2 XPS spectra of (a) wide scan and (b) narrow scan of Ni orbital (after ion-beam etching of surface carbon).

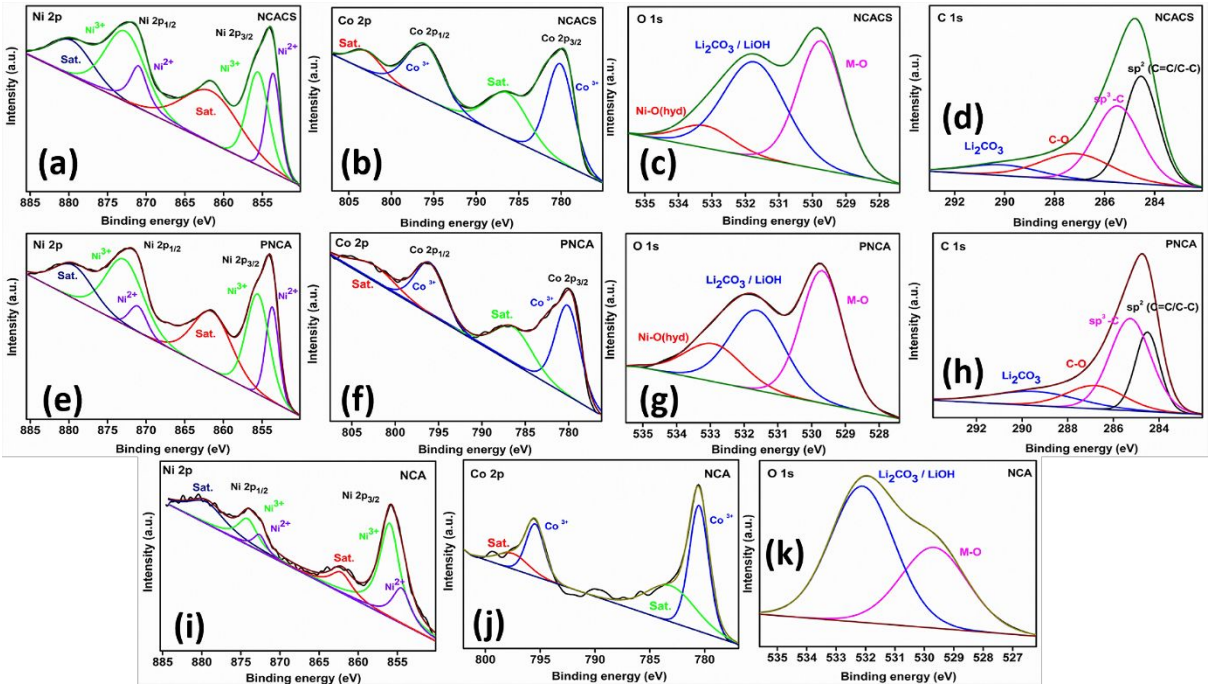

Figure. S3 Deconvoluted XPS spectra of Ni 2p, Co 2p, O 1s and C 1s orbitals of NCACS (a-d), PNCA (e-h) and NCA (i-k) respectively.

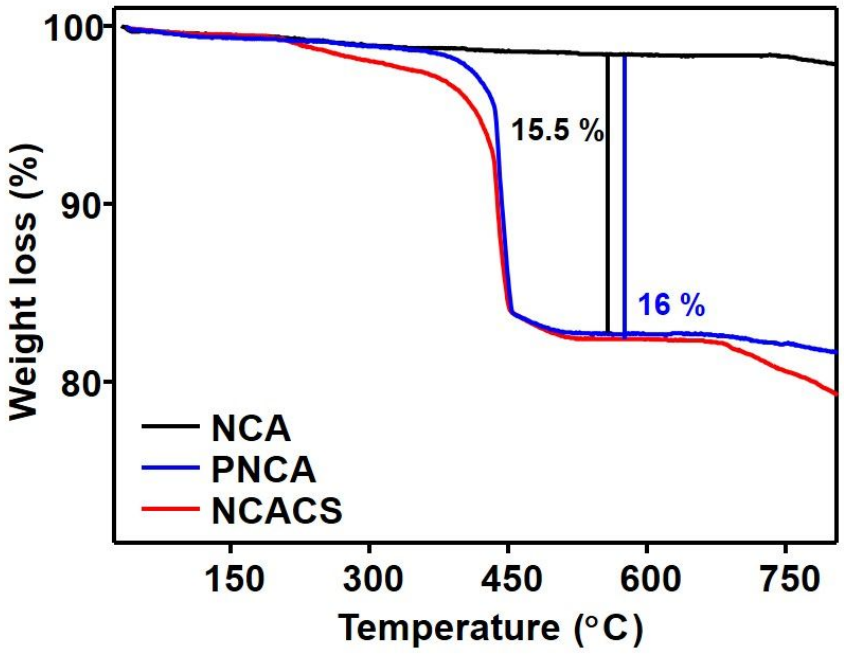

Figure. S4 Thermo-gravimetric curves of neat NCA, carbon coating obtained by physical blending (PNCA) and mechanical force reinforced mechano-fusion (NCACS) in an oxygen atmosphere.

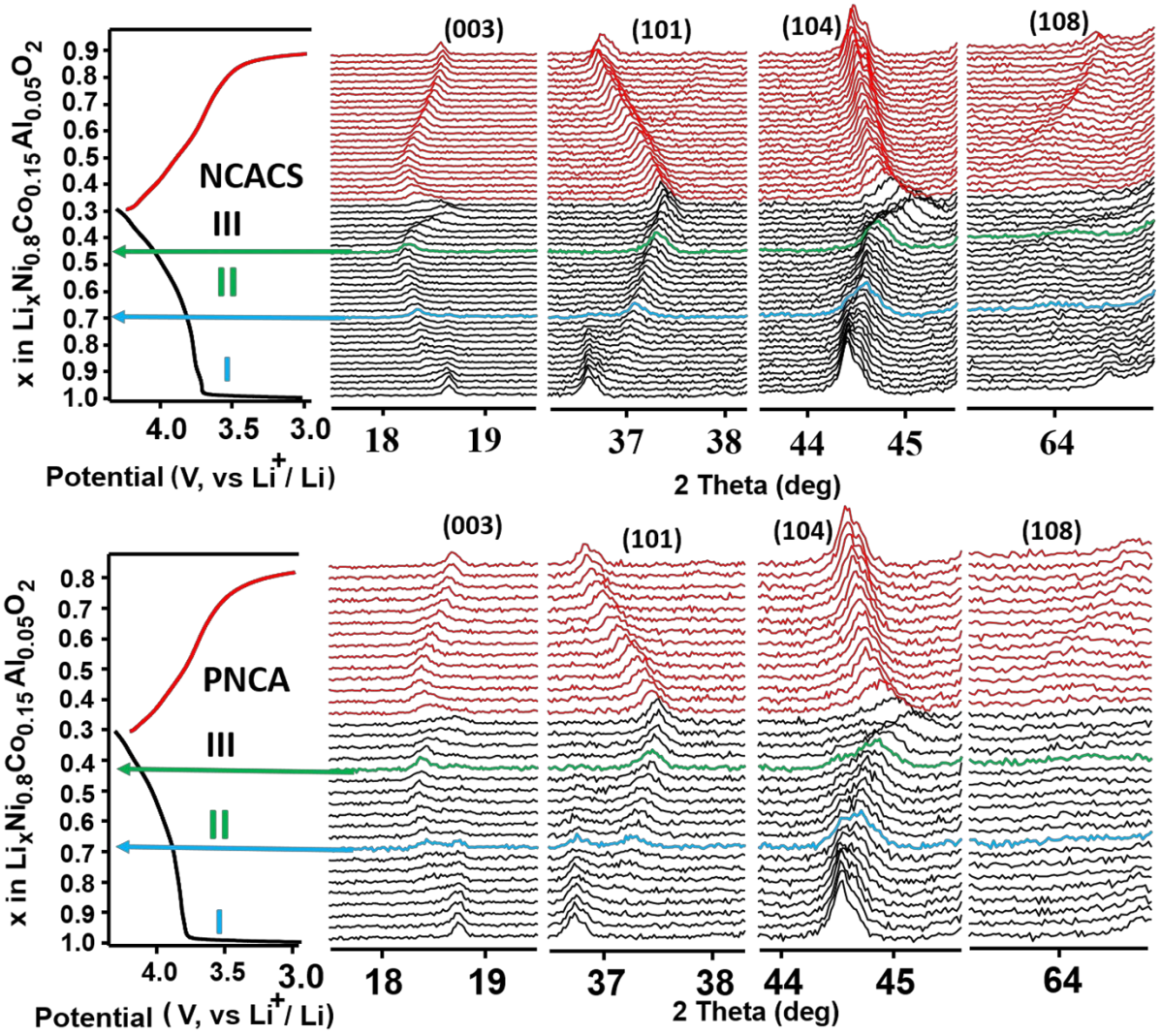

Figure. S5 Selected region diffraction pattern for the evolution of new peak observed in *operando* XRD concerning to lithium ion concentration of NCACS and PNCA ($LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$) during the first cycle at 0.5 C.

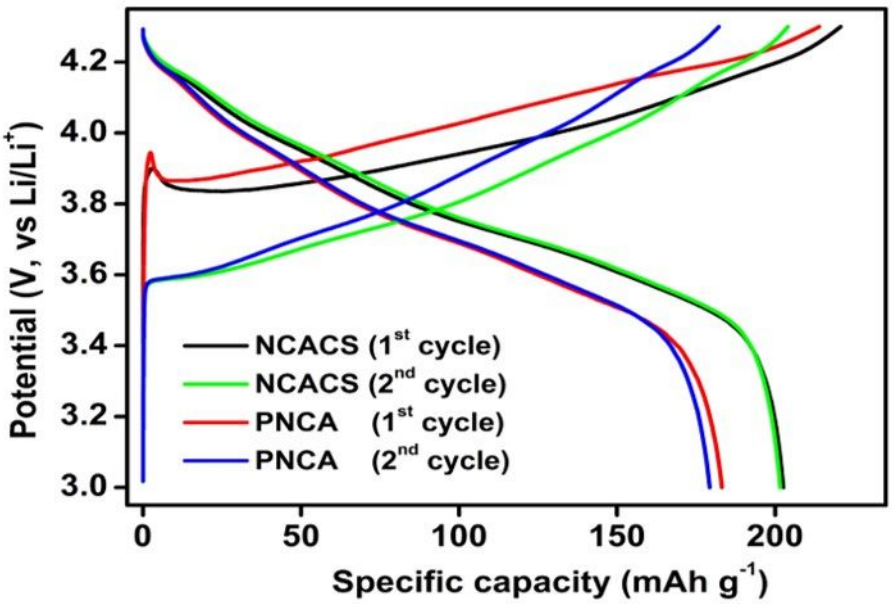

Figure. S6 Galvanostatic cycling curves for the first and second cycles of NCACS and PNCA vs. Li^+ / Li at 0.1 C.

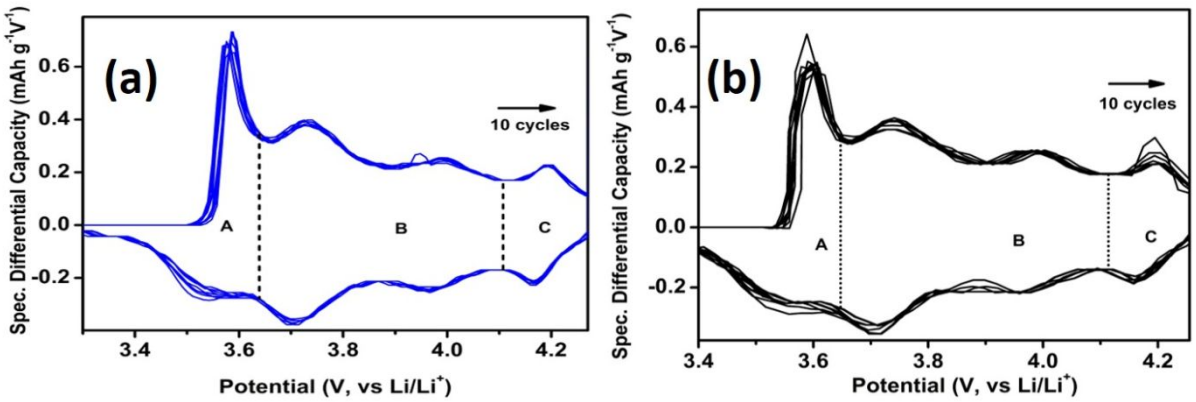

Figure. S7 Differential Specific capacity as a function of voltage (dQ/dV vs V g^{-1}) for (a) NCACS and (b) PNCA calculated at 0.1 C, respectively.

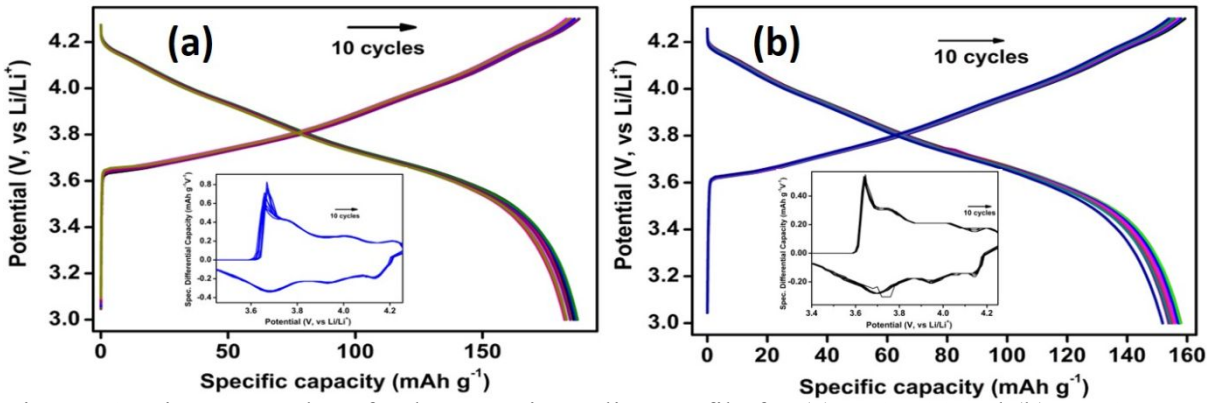

Figure. S8 First ten cycles of galvanostatic cycling profile for (a) NCACS and (b) PNCA at 0.5 C (inset: differential specific capacity as a function of voltage (dQ/dV vs V g^{-1})).

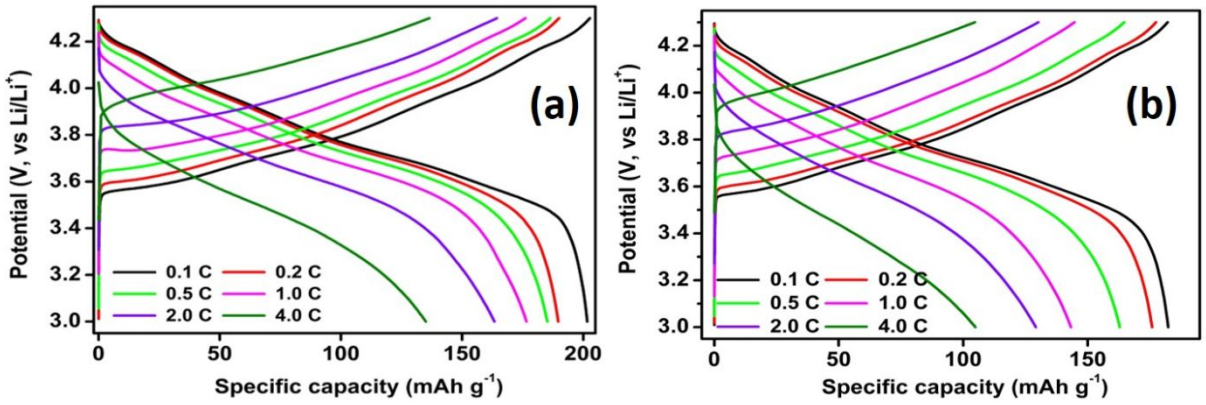

Figure. S9 Galvanostatic cycling profile at various C rates for (a) NCACS and (b) PNCA.

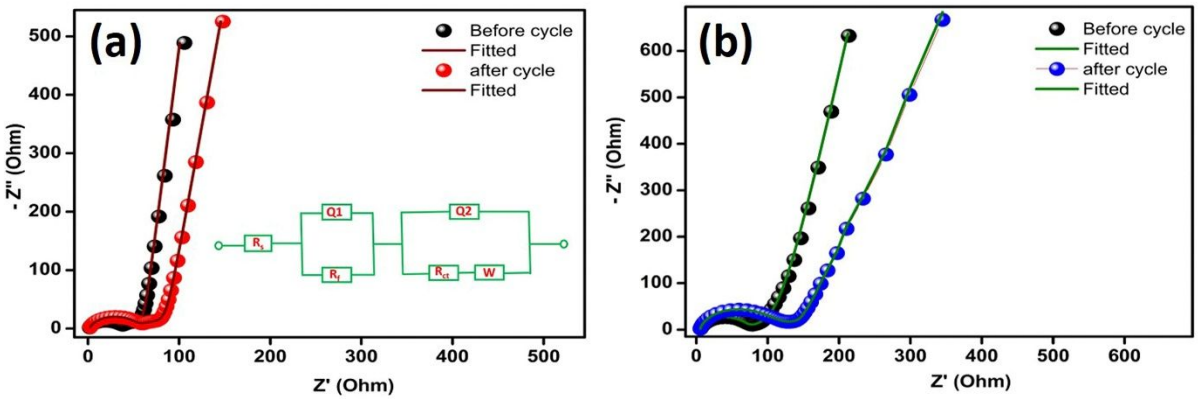

Figure. S10 Electrochemical impedance spectra and fitted results for (a) NCACS and (b) PNCA before and after cycling (inset: generated Randle's equivalent circuit).

Table S1. Comparison of electrochemical performance of different NCA materials

References

1. Chen, Y.; Li, P.; Zhao, S.; Zhuang, Y.; Zhao, S.; Zhou, Q.; Zheng, J. Influence of integrated microstructure on the performance of $LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$ as a cathodic material for lithium ion batteries. *RSC Adv.* ***2017****, 7*(46), 29233–29239.

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S.No	Material	Specific discharge capacity (mAh g^{-1})	Rate	Remarks
1.	Concentration gradiant NCA	165	1 C	[1]
2.	Na-doped NCA	160	1 C	[2]
3.	N/P doped NCA	170	1 C	[3]
4.	$LiAlO_2$ coated NCA	170	1 C	[4]
5.	Spherical precursor- NCA	180	0.5 C	[5]
6.	LBO-coated NCA	163	1 C	[6]
7.	Cation mixing regulated NCA	163	1 C	[7]
8.	NCA microrods	181	0.5 C	[8]
9.	**NCACS**	**186**	**0.5 C**	
		178	**1 C**	
	PNCA	**161**	**0.5 C**	**Present study**
		146	**1 C**	

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