Supporting Information

Activation of residual sodium via in-situ neutralization toward practical layered oxides

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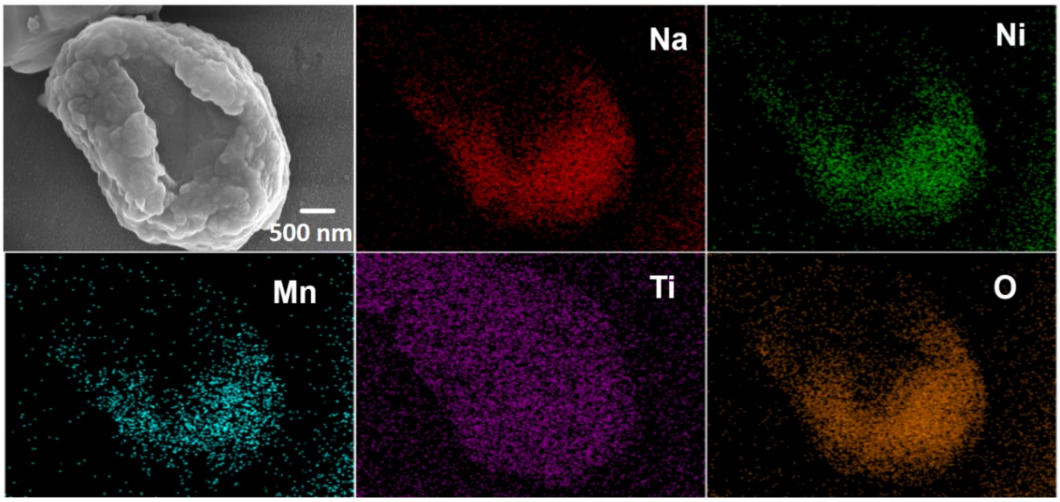

Figure. S1. EDS mapping images of P2-NMT.

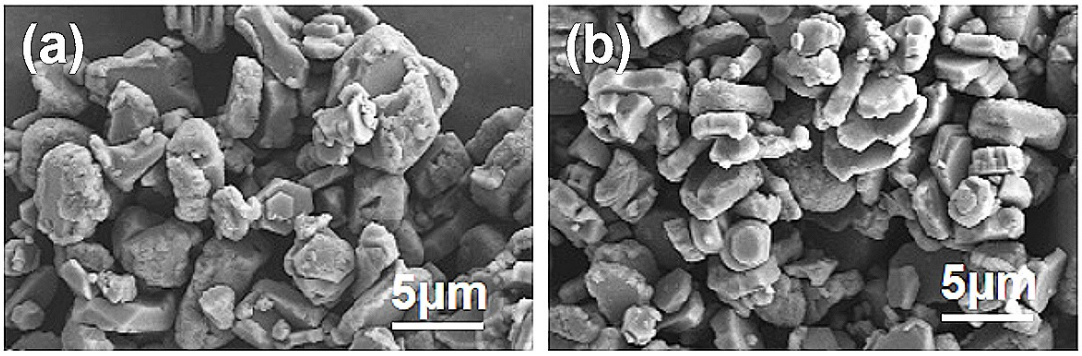

Figure. S2. SEM images of (a) NMT and (b) NMT@IDA.

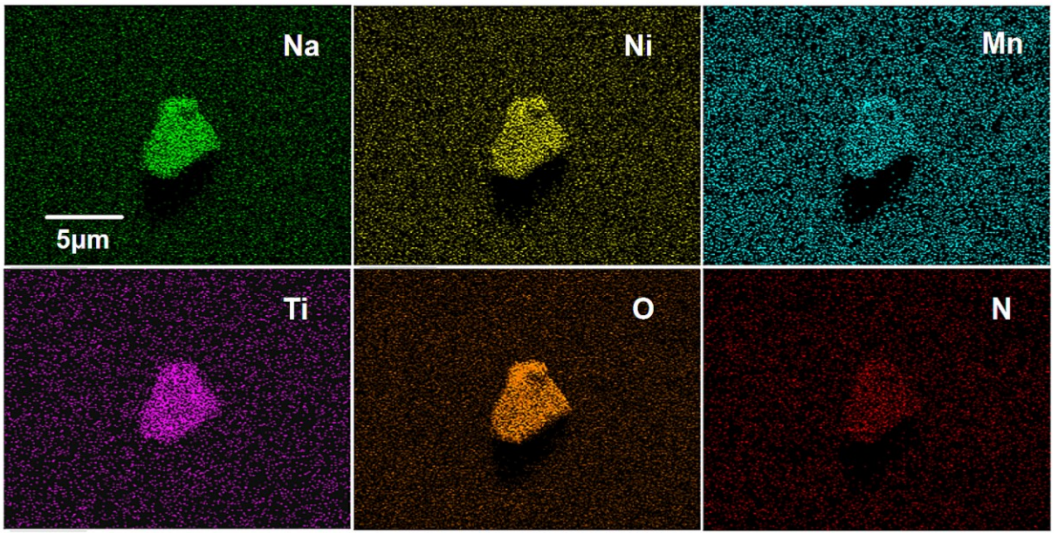

Figure. S3. EDS mapping images of P2-NMT@IDA.

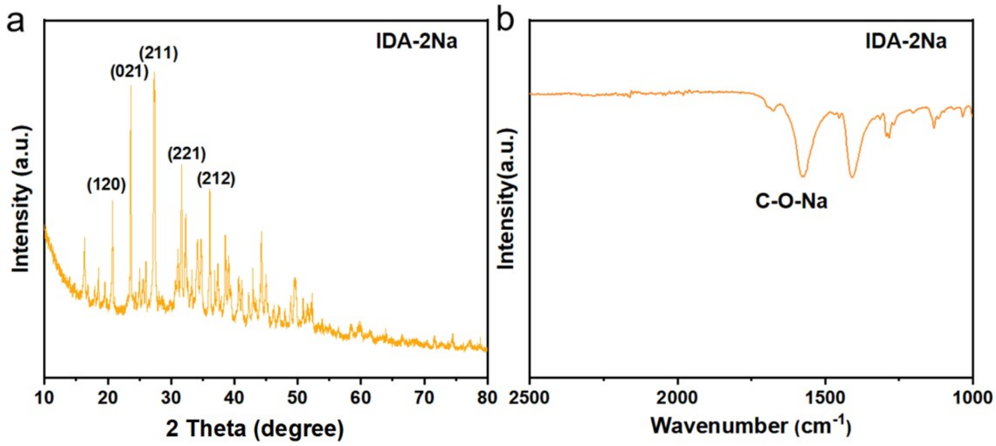

Figure. S4. (a) XRD patterns of IDA-2Na, (b) Fourier transform infrared spectroscopy (FTIR) patterns of IDA-2Na.

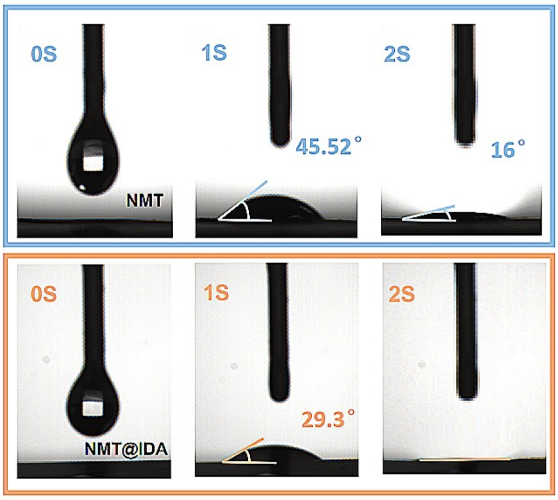

Figure. S5. The wetting snapshot of electrolyte droplets of (a) P2-NMT and (b) P2-NMT@IDA electrodes.

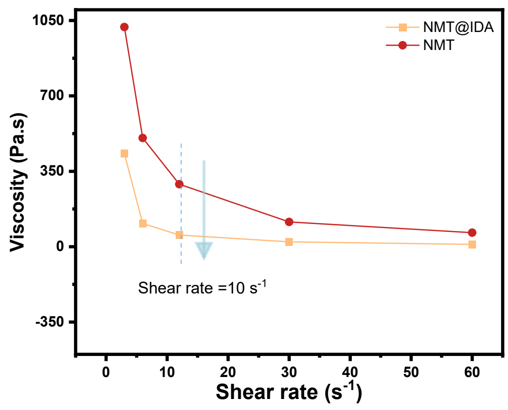

Figure. S6. Oscillatory shear rheology of the electrode slurry.

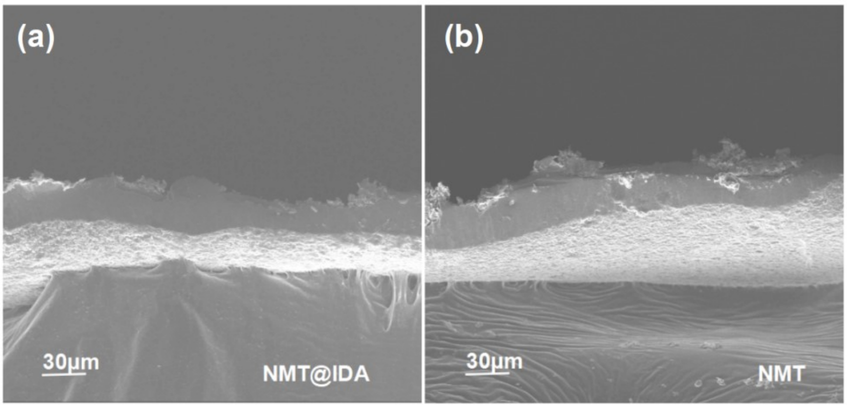

Figure. S7. SEM analyses for the morphology of (a) P2-NMT and (b) P2-NMT@IDA electrodes, in which show the cross-sectional images.

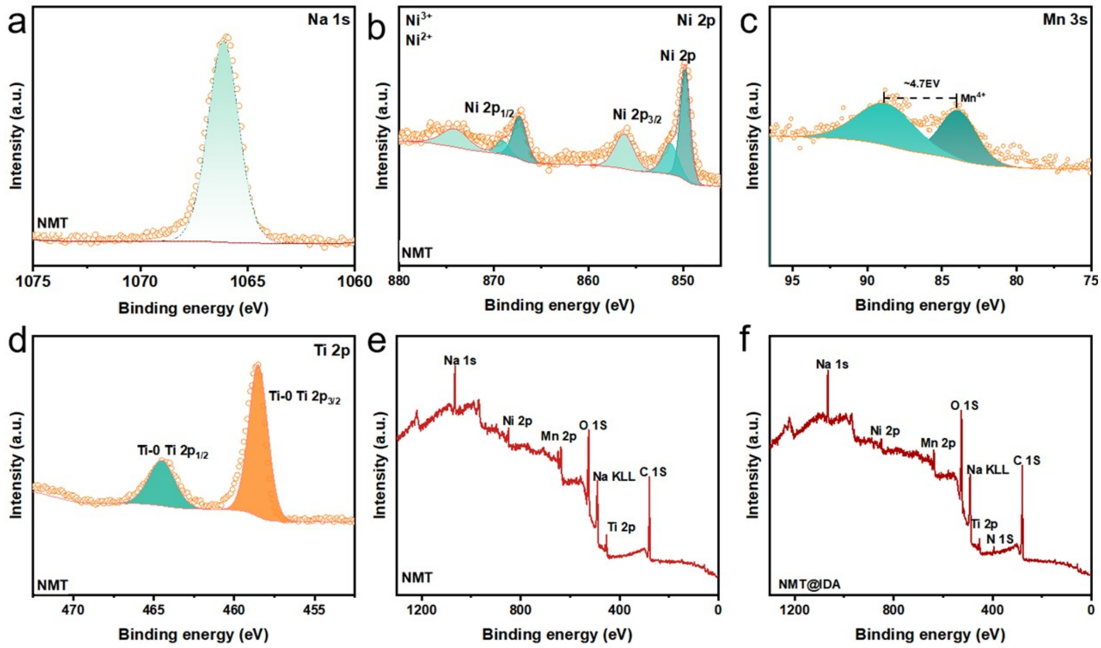

Figure. S8. High-resolution (a) Na 1s, (b) Ni 2p, (c) Mn 3s, (d) Ti 2p XPS spectrum of NMT sample. High-resolution (d-f) XPS full spectrum of NMT and NMT@IDA sample.

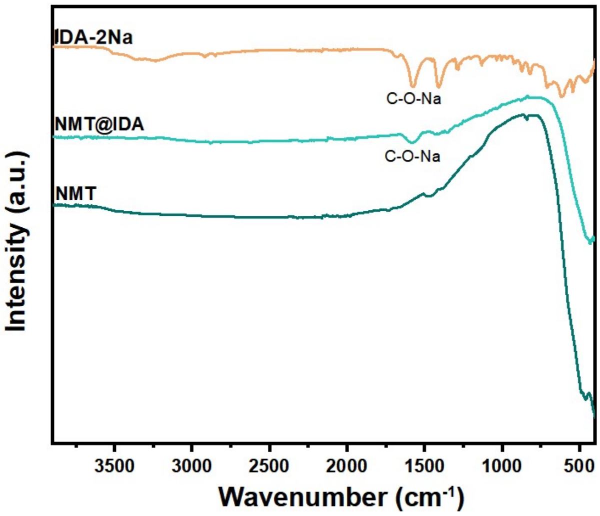

Figure. S9. FTIR spectra for P2-NMT, P2-NMT@IDA and IDA-2Na sample.

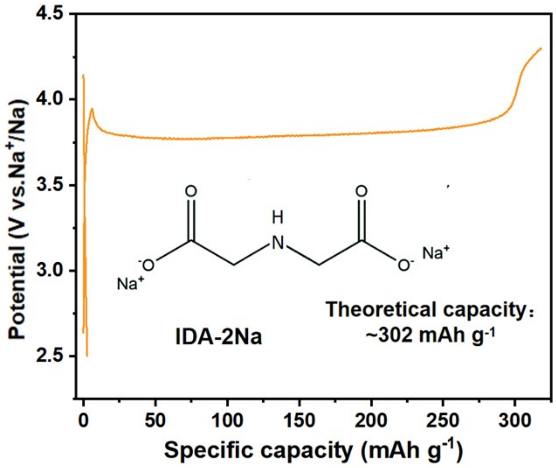

Figure. S10. Electrochemical properties of IDA-2Na at 0.1 C.

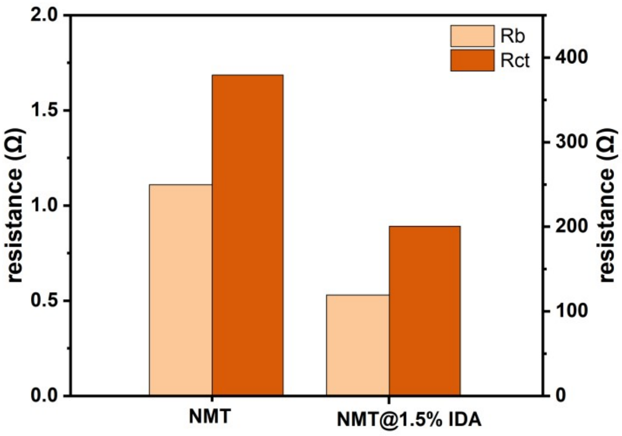

Figure. S11. The value of impedance parameters R_b and R_{ct} that received by fitting the Nyquist plots of P2-NMT and P2-NMT@IDA cathodes.

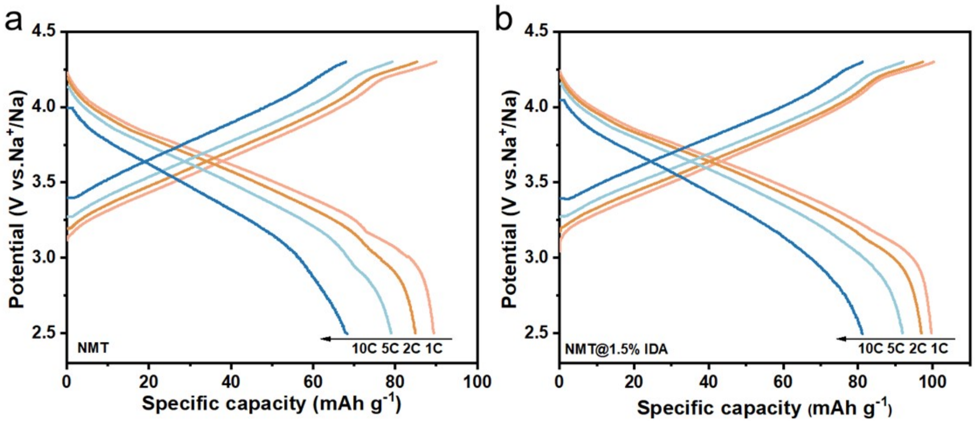

Figure. S12. The rate capabilities of P2-NMT and P2-NMT@IDA at different current densities. The charge/discharge curves of (a) P2-NMT||Na and (b) P2-NMT@IDA||Na cells at rates of 1, 2, 5, and 10 C.

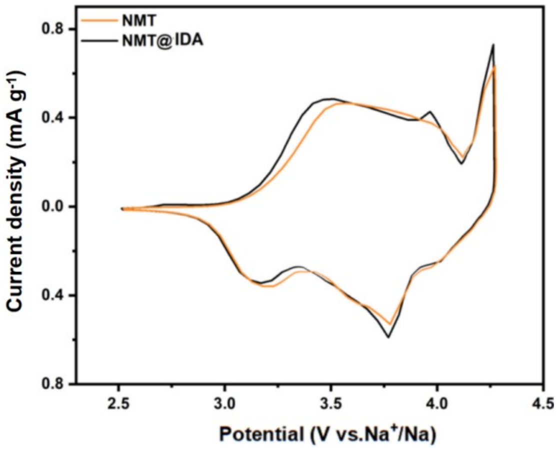

Figure. S13. CV curves of half-cells at 0.1 mV s^{-1}.

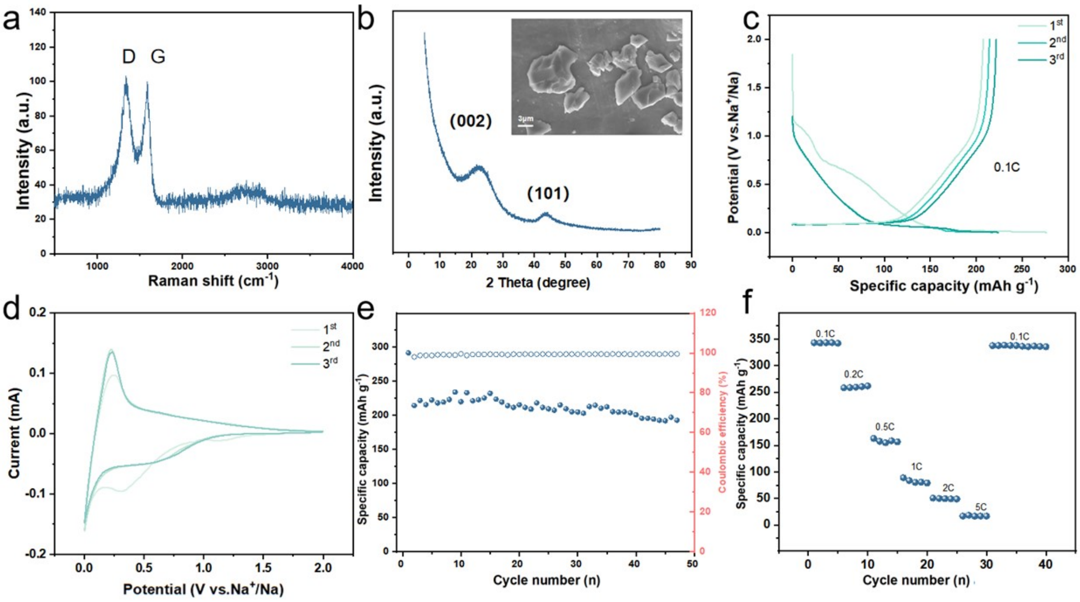

Figure. S14. Characterization of the structure and properties of hard carbon materials. (a) Raman spectrum. (b) SEM image and XRD pattern. (c) Charge and discharge profiles at 0.1 C in the voltage range of 0.01 to 2.0 V. (d) CV curves at 0.1 mV s^{-1}. (e) Cycle performance and (f) rate performance.

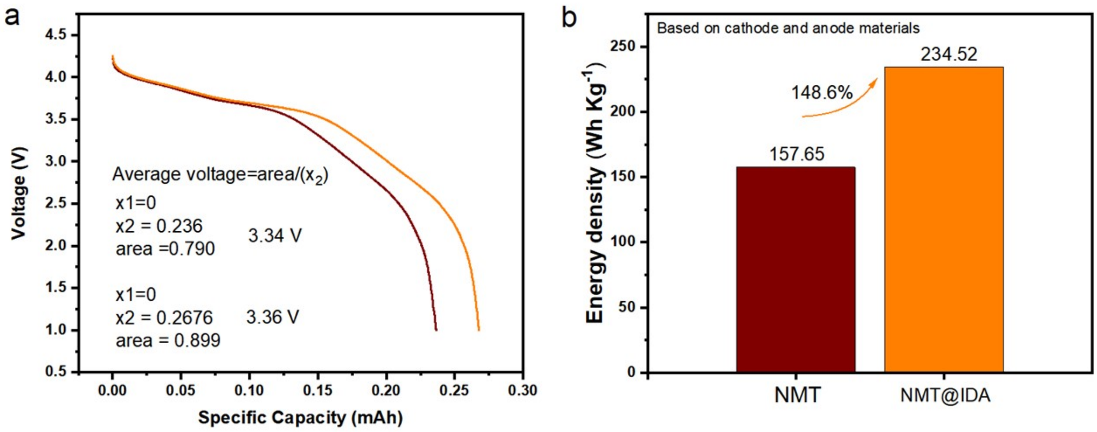

Figure S15. (a) Comparison of charge/discharge profiles energy density of NMT and NMT@IDA in full cells. (b) Integral results of the discharge profiles and the calculation of the corresponding average voltage.

Table S1. Lattice parameters of P2-NMT and P2-NMT@IDA samples.

Sample	a /Å	b /Å	c /Å	α	β	γ
$Na_{2/3}Ni_{1/3}Mn_{1/3}Ti_{1/3}O_2$	2.924025	2.924025	11.159918	90°	90°	120°
$Na_{2/3}Ni_{1/3}Mn_{1/3}Ti_{1/3}O_2$@IDA	2.924835	2.924835	11.162878	90°	90°	120°

Table S2. Rietveld refinement of XRD data for P2-NMT.
NMT. Space group P63/mmc.
a=b=2.924025 Å; c=11.159918 Å;
$\alpha=\beta=90°$; $\gamma=120°$; V=82.633 Å^3.
R_{wp}=4.64%; R_p=3.40%.

Atom	x	y	z	Occupancy
Na_f	0.00000	0.00000	0.25000	0.192
Na_e	0.33333	0.66667	0.75000	0.475
Mn	0.00000	0.00000	0.00000	0.333
Ni	0.00000	0.00000	0.00000	0.333
Ti	0.00000	0.00000	0.00000	0.333
O	0.33333	0.66667	0.08391	1.000

Table S3. Rietveld refinement of XRD data for P2-NMT@IDA.
NMT@IDA. Space group P63/mmc.
a=b=2.924835 Å; c=11.162878 Å;
$\alpha=\beta=90°$; $\gamma=120°$; V=82.701 $Å^3$.
$R_{wp}=5.07\%$; $R_p=3.64\%$;

Atom	x	y	z	Occupancy
Naf	0.00000	0.00000	0.25000	0.192
Nae	0.33333	0.66667	0.75000	0.475
Mn	0.00000	0.00000	0.00000	0.333
Ni	0.00000	0.00000	0.00000	0.333
Ti	0.00000	0.00000	0.00000	0.333
O	0.33333	0.66667	0.07980	1.000

Table S4. ICP-OES results for P2-NMT and P2-NMT@IDA.

Sample	CHNS Weight (mg) by ICP-AES	Na (%) by ICP-AES	Ni (%) by ICP-AES	Ti (%) by ICP-AES
$Na_{2/3}Ni_{1/3}Mn_{1/3}Ti_{1/3}O_2$	11.3	13.9	19.62	14.6
$Na_{2/3}Ni_{1/3}Mn_{1/3}Ti_{1/3}O_2$@IDA	11.8	14.5	20.1	15.9

Table. S5 Quantitative analysis of surface residual alkali in the pristine NMT sample.

Sample	NaOH content (wt%)	Na_2CO_3 content (wt%)
NMT	0.24 wt%	0.43 wt%

Note S1

In this work, calculation of Stoichiometric IDA Requirement can be calculated based on the following two equations:

$$\mathbf{IDA + 2NaOH \rightarrow IDA - 2Na + 2H_2O} \quad (1)$$

$$\mathbf{2IDA + Na_2CO_3 \rightarrow 2IDA - 2Na + H_2O + CO_2\uparrow} \quad (2)$$

Calculations of IDA Requirement are based on the equations.

The formula is:

$$\text{IDA (\%)} = \frac{133.10}{2 \times M_{NaOH}} \times \text{NaOH content (wt\%)} + \frac{2 \times 133.10}{M_{Na_2CO_3}} \times Na_2CO_3 \text{ content (wt\%)},$$

where M_{NaOH} and M_{Na2CO3} are the relative molecular mass of NaOH and Na_2CO_3, respectively.

Specific calculation process based on the Table. S4:

1. IDA required to neutralize NaOH

Molar mass: IDA = 133.10 g • mol^{-1}, NaOH = 40.00 g • mol^{-1}.

Stoichiometry: 1 mol IDA reacts with 2 mol NaOH.

Mass of IDA needed to neutralize 0.24 g NaOH per 100 g cathode:

133.102×40.00×0.24≈0.40 g per100 g cathode=0.40wt%

2. IDA required to neutralize Na_2CO_3

Molar mass: Na_2CO_3 = 105.99 g • mol^{-1}.

Stoichiometry: 2 mol IDA reacts with 1 mol Na_2CO_3.

Mass of IDA needed to neutralize 0.43 g Na_2CO_3 per 100 g cathode:

2×133.10105.99×0.43≈1.08 g per 100 g cathode=1.08 wt%

3. Total IDA required for complete neutralization:

0.40wt%+1.08wt%=1.48wt%

4. Practical application:

In the actual slurry environment, factors such as solubility of IDA in NMP, dispersion homogeneity, and surface accessibility may affect the reaction efficiency. A slight excess

(1.5 wt%) helps ensure complete neutralization and uniform coating formation.

Performance optimum: As shown in Fig. 4e-f, 1.5 wt% IDA delivered the best overall electrochemical performance, indicating that this loading balances complete alkali conversion with the formation of a functional interfacial layer without introducing excessive porosity or impedance.