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Accelerating the Activation of Li_2MnO_3 in Li-Rich High-Mn Cathodes to Improve Its Electrochemical Performance

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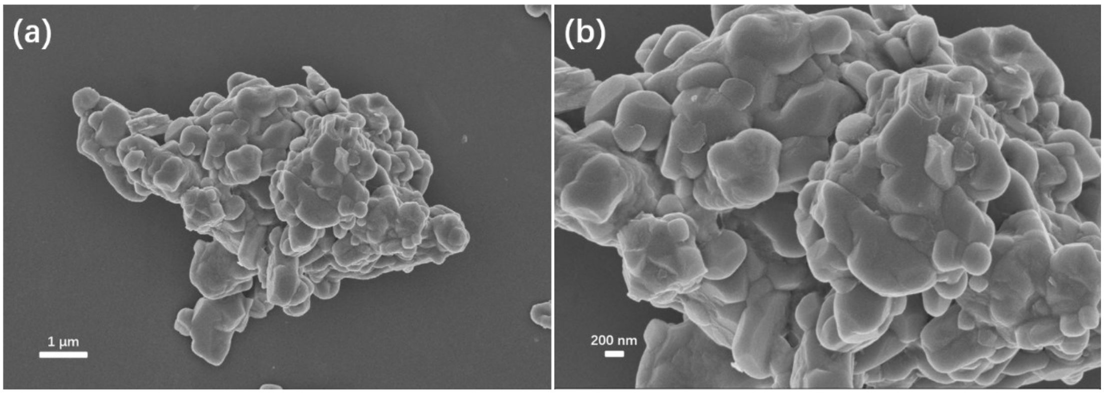

Figure S1. The SEM images of NHM-811 without special appearance was synthesized by solid-state reactions.

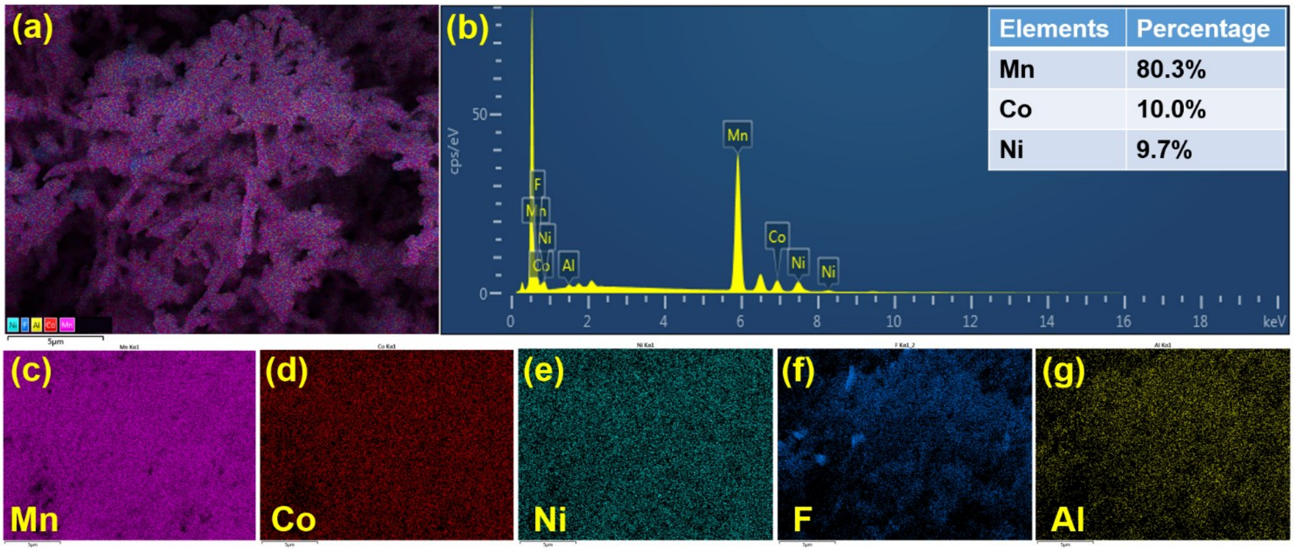

Figure S2. The SEM images of and the EDS mapping of RHM-811@3%LAF.

Fig. S2 show the Mn, Co, Ni, Al, and F homogeneously distribute in RHM-811@3%LAF. The concentration ratio of Mn, Co, and Ni is close to 8:1:1, which is consistent with the previous ICP-AES results.

Table S1. Result of inductively coupled plasma atomic emission spectroscopy

	Li	Mn	Ni	Co
mg/L	5.588	21.059	2.697	2.626
mol/L	0.8051	0.3833	0.0460	0.0445
Mole rate	1.700	0.809	0.097	0.094

Table S2. Detail of 1st Charge capacity of all samples

1st Charge capacity	NHM-811	RHM-811	RHM-811 @1%LAF	RHM-811 @2%LAF	RHM-811 @3%LAF
below 4.5 V ($mAh\cdot g^{-1}$)	36.4	65.9	80.6	73.3	68.8
above 4.5 V ($mAh\cdot g^{-1}$)	70.8	208.1	238.6	241.3	237.4
Ratio of above 4.5 V (%)	66.0	75.3	74.7	76.7	77.5

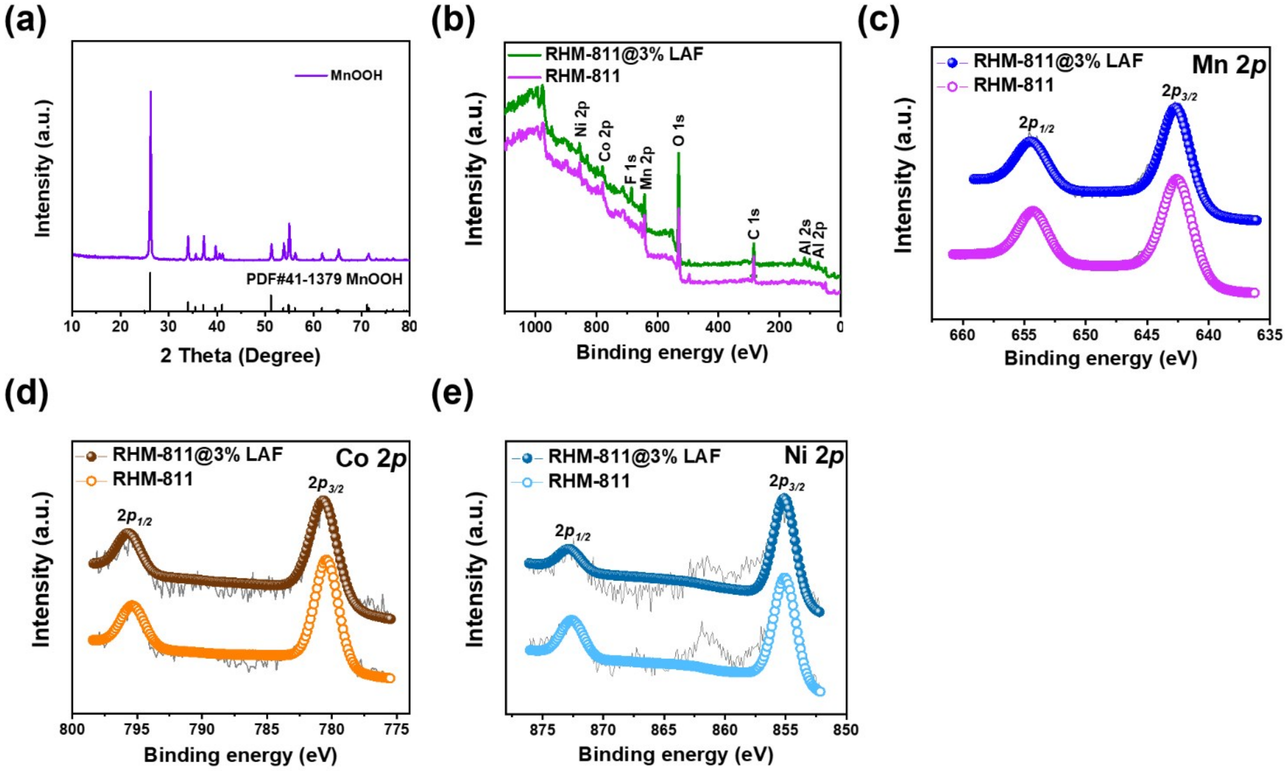

Fig.S3 (a) The survey scan and (b) Al 2p, (c) F 1s, (d) Mn 2p, (e) Co 2p, (f) Ni 2p narrow scan of X-ray photoelectron spectroscopy (XPS) of the RHM-811 and RHM-811@3%LAF.

Among them, the two well-defined peaks at 74.1 eV and 685.15 eV can be observed in the spectra, attributed to the Al^{3+} 2p and F^{-} 1s (Fig.S3b); Furthermore, the chemical state of Ni, Co, Mn element in the samples was analyzed by XPS narrow scan of its 2p core-level was displayed in Fig.3c-e. According to the fitting results, the peaks of RHM-811 at 654.2/642.5, 780.4/795.3, 872.6/855.1 eV corresponding to typical Mn^{4+}, Co^{3+}, Ni^{2+} $2p_{1/2}/2p_{3/2}$ peaks, respectively; Meanwhile, the Mn^{4+}, Co^{3+}, Ni^{2+} $2p_{1/2}/2p_{3/2}$ peaks of RHM-811@3%LAF at 654.3/642.6, 780.6/795.6, 872.8/855.1 eV, respectively. Comparing the above results, it can be seen that there is almost no change in the 2p peak position of Ni, Co, and Mn.

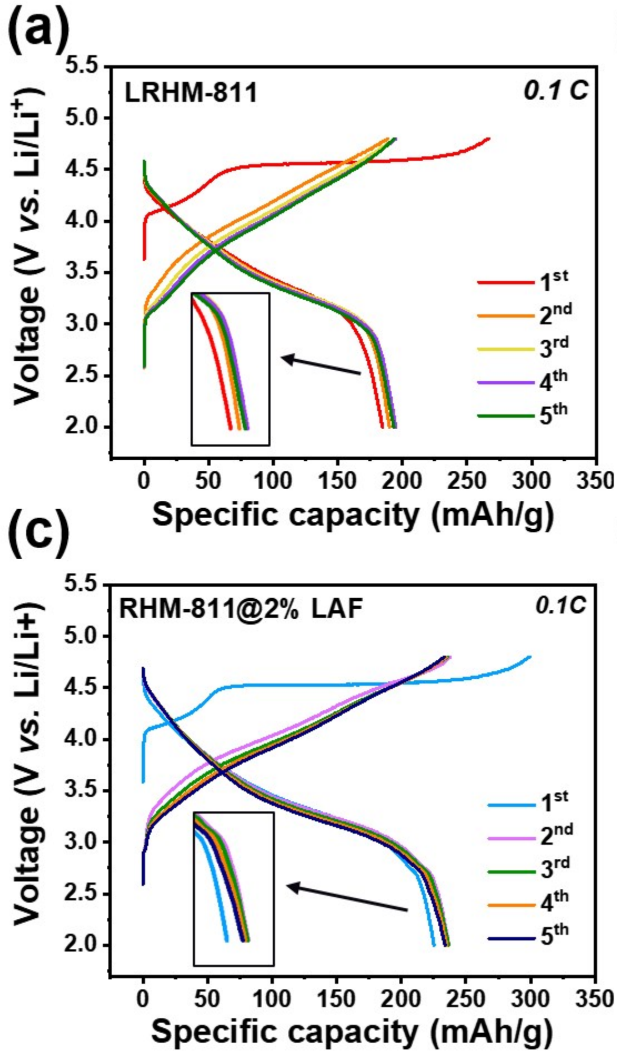

Figure S4. The first 5 discharge curves of (1) RHM-811, (2) RHM-811@1%LAF, (3) RHM-811@2% LAF, (4) RHM-811@3% LAF at 0.1 C between 2.0 and 4.8 V.

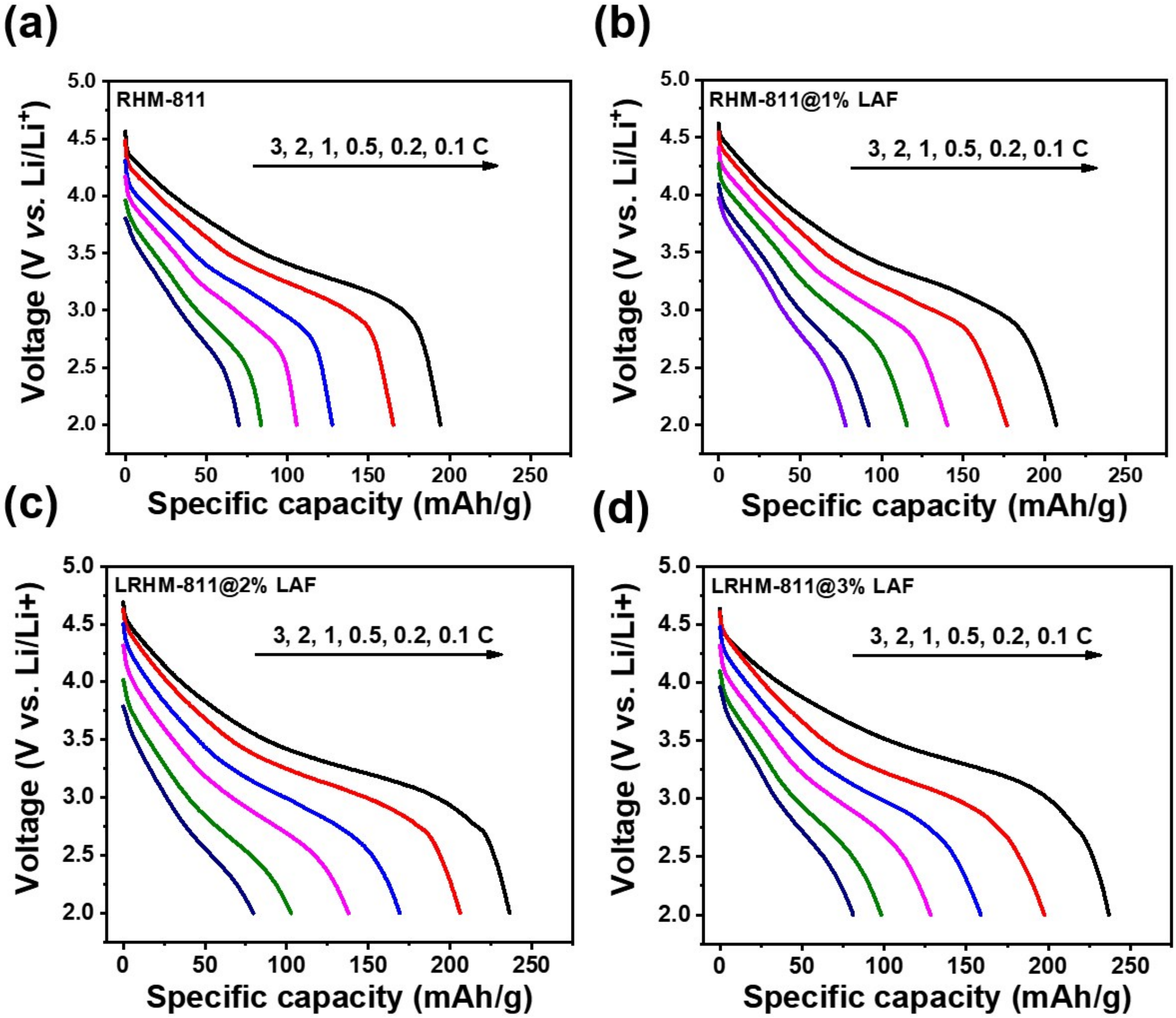

Figure S5. The discharge curves of (a) RHM-811, (b) RHM-811@1%LAF, (c) RHM-811@2% LAF, (d) RHM-811@3% LAF at 0.1, 0.2, 0.5, 1.0, 2.0, 3.0 C between 2.0 and 4.8 V.

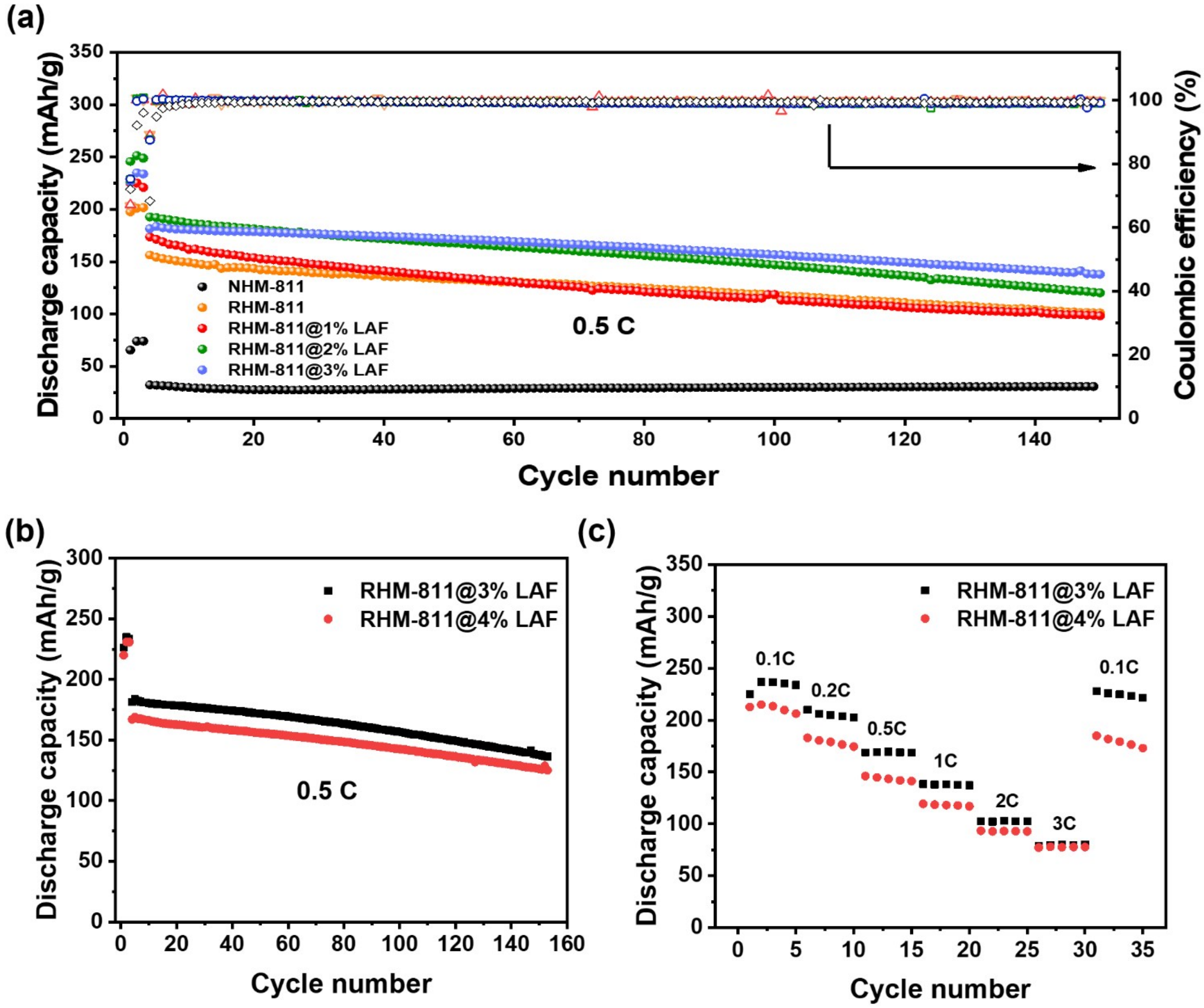

Figure S6. The 0.5C cycle and rates performance.

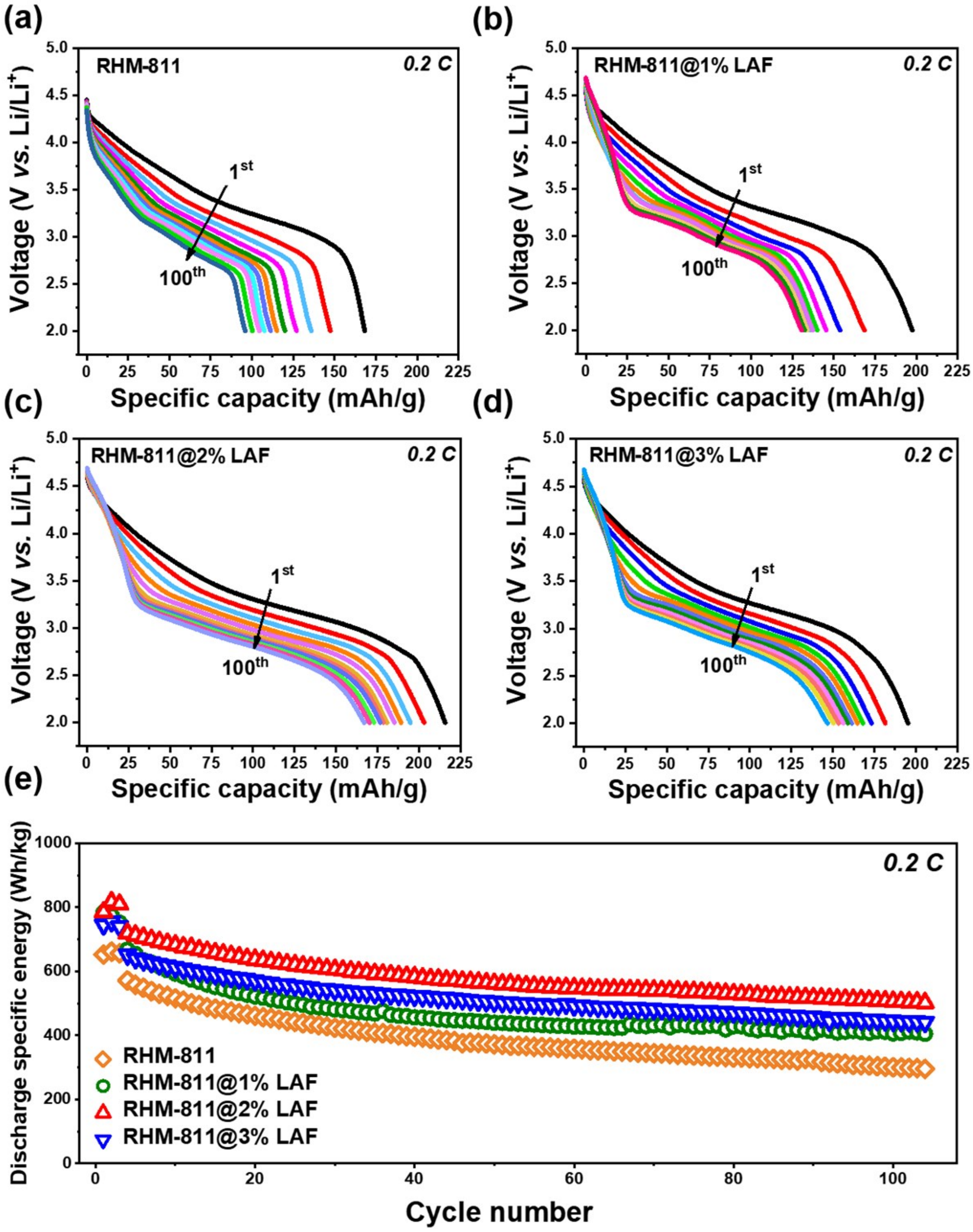

Figure S7. The voltage drop curve and the energy density as a function of the number of cycles.

Figure S8. Photograph of 2032 cells being tested in digital high-low temperature test chamber.

NOTE1: The calculation method of the Li^+ diffusion potential barrier E_a

The relationship between the $D_{Li}$and temperature T can be expressed by the Arrhenius equation:

$$D_{Li} = D_0 e^{-\frac{E_a}{k_B T}}$$

The D_0 is the diffusion constant, and E_a is the diffusion potential barrier, which depend on the composition and structure of materials, independent of temperature. *k* is the B`oltzmann constant (k = 1.3806505(24) × 10^{-23} J/K). We take the natural logarithm of both sides of Arrhenius equation and get the equation:

$$lnD_{Li} = lnD_0 - \frac{E_a}{k_B T}$$

NOTE2: The calculation method of D_{Li}

In the equivalent circuit (Fig.7g), R_s and R_{ct} represent the ohmic resistance (caused by electrolyte) and the charge transfer resistance at the electrolyte/electrode, which corresponds to the high-frequency intercept at the $Z^{'}$ axis. CPE represents capacitance. W is Warburg element. In addition, the lithium ion diffusion coefficient (D_{Li}) was related to the Li^+ diffusion process within the electrodes, which can be calculated from the Nyquist plot in low-frequency section, according to the following equation:

$$Z^{'} = R_{ct} + R_S + \sigma\omega^{-1/2} \tag{1}$$

$$D_{Li} = \frac{R^2T^2}{2A^2n^4F^4{C_{Li}}^2\sigma^2} \tag{2}$$

where ω is the angular frequency, R is the gas constant (8.314 J/(mol·K)), A is the surface area of the electrode(1.13 cm^2), T is the absolute temperature, F is the Faraday constant (96485.34 C/mol), n is the number of electrons transferred in the half-reaction for the redox couple, and C_{Li} is the concentration of lithium ion (1*10^{-3} mol/cm^{-3}). σ is the Warburg factor that is relative to $Z^{'}$ *vs.* $\omega^{-1/2}$

and can be obtained from the slope of the lines in Fig. 7h. The lithium ion diffusion coefficients are calculated by equation (2).

Table S3. The fitted results from EIS.

	RHM-811	RHM-811@3%LAF
R_s / Ω	2.055	3.895
R_{ct} / Ω	1280	230
D_{Li} / $cm^2\ s^{-1}$	6.92×10^{-15}	1.11×10^{-14}