Supporting Information

Li-In alloy anode and Nb\textsubscript{2}CT\textsubscript{X} artificial solid-electrolyte interphase for practical Li metal batteries.

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Fig. S1. Li-In phase diagram.

Fig. S2. XRD patterns of Li-In alloy anode after Li stripping or plating.

Fig. S3. BSE, EDX element mapping, FIB-SEM cross-sectional image of Li-In alloy anode after Li stripping.

Fig. S4. Molecular structure of Li$_{13}$In$_3$ unit cell and Li diffusion pathway on (311) surface of Li$_{13}$In$_3$.

Fig. S5. SEM image and corresponding EDX element mapping image of Nb$_2$CT$_X$ MXene and Nb$_2$AlC MAX phase.

Fig. S6. XRD patterns of Nb$_2$AlC and Nb$_2$CT$_X$.

Fig. S7. Wide scan XPS spectra of Nb$_2$CT$_X$ MXene and Nb$_2$AlC MAX phase.

Fig. S8. Optical image of manufacturing process and SEM images of Nb$_2$CT$_X$ Li-In anode.

Fig. S9. XRD profile of Nb$_2$CT$_X$ Li-In anode.

Fig. S10. Practical areal capacity measurement of Nb$_2$CT$_X$ Li-In anode.

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Fig. S12. Galvanostatic Li stripping/plating voltage profile.

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Fig. S14. AC impedance measurement for Li|NCM 811, Li-In|NCM 811, Li-In Nb$_2$CT$_X$|NCM 811.

Fig. S15. Li-In foil thickness and further roll-pressing results.

Table S1. The calculation for termination group formation energy and Li binding energy on the Nb$_2$CT$_X$ surface.

Table S2. Li-In foil thickness before and after roll-pressing.
Fig. S1. Li-In phase diagram. The red dashed vertical line represents 80 wt% of Li (98.51 at % of Li) and 20 wt % of In (1.49 at % of In) composition. At the designed composition, Li-In alloy consists of 74.68 wt % of Li and 25.32 wt % of Li$_{13}$In$_3$. 
Fig. S2. XRD patterns for Li-In alloy anode after a) 4 mAh cm\(^{-2}\) of Li stripped, b) 4 mAh cm\(^{-2}\) Li deposited back to a), c) 37.5 mAh cm\(^{-2}\), d) 39.0 mAh cm\(^{-2}\), e) 39.5 mAh cm\(^{-2}\), f) all of Li stripped from Li-In alloy anode. Phase transition from Li\(_{13}\)In\(_3\) to In was observed after 37.5 mAh cm\(^{-2}\) of Li stripped (Li\(_{13}\)In\(_3\) → Li\(_2\)In → LiIn → In). XRD peak of Li, Li\(_{13}\)In\(_3\), Li\(_2\)In, LiIn, and In notice with the filled circle, empty-triangle, filled-diamond, empty-square, and empty-star shape, respectively.
Fig. S3. a) SEM BSE mode micrographs of the Li-In alloy anode after 4 mAh cm$^{-2}$ galvanostatic stripping of Li using Li-In|Li-In symmetric cell. Bright particles are Li-In alloy (Li$_{13}$In$_3$), and the dark background is pristine Li. b) Corresponding EDX element mapping image for In (red). c) FIB cross-section image of Li-In after Li stripping with the same conditions as a). In the electrode's upper region, most of the Li was stripped, and only the light-colored Li$_{13}$In$_3$ remains, showing an irregular porous structure, and the remained Li shows dark in the lower part of the image.
Fig. S4. a) The optimized crystal structure of Li$_{13}$In$_3$. The space group is the cubic Fd$^3$m with the lattice parameters $a = b = c = 13.55$ Å and $\alpha = \beta = \gamma = 90^\circ$. Visualization of the Li migration on the (311) surface of the Li$_{13}$In$_3$ along b) pathway 1 (Li$_{13}$In$_3^\bullet$), c) pathway 2 (Li$_{13}$In$_3^{**}$). The first configuration is the initial state of the diffusion process that Li adsorbed on the most stable adsorption site on the (311) of L$_{13}$In$_3$ and considered as 0 eV. The adsorbed Li migrates following its stable energetic position, and each image shows the Li when it was located at a local minimum. Values below each image are relative energy calculated with respect to the energy at the initial state. Blue and yellow spheres are In and Li of L$_{13}$In$_3$, and the red sphere is the migrating Li adatom.
Fig. S5. SEM image and EDX element mapping images of a) Nb$_2$CT$_X$ (MXene) and b) Nb$_2$AlC (MAX phase). EDX element mapping images in both contents correspond with Nb L (purple), C K (red), Al K (yellow), F K (blue), O K (oxygen) peak.
Fig. S6. XRD profiles of Nb$_2$AlC (bottom, black) and Nb$_2$CT$_X$ (top, red). Marks on the graph correspond to the major XRD peaks Nb$_2$AlC (black triangle), Nb$_2$CT$_X$ (red star). After the synthesis, significant changes of several peaks indicate the transformation of MAX phase to MXene; the (103) peak of Nb$_2$AlC, around 2$\theta$ of 39°, disappears after synthesis, and the (002) peak of Nb$_2$AlC corresponding to 2$\theta$ of 12.9° shifts to a lower angle. Disappeared (103) peak represents that Al layer was removed, and shifted (002) peak means that inter-layer space of Nb$_2$CT$_X$ was increased.
Fig. S7. a) Wide scan XPS spectra of Nb$_2$CT$_x$ (MXene) and Nb$_2$AlC (MAX phase), b) High resolution XPS spectra of Nb$_2$AlC (black) and Nb$_2$CT$_x$ (red) at Al 2p region.
Fig. S8. Preparation and morphologies of the Nb$_2$CT$_X$ Li-In anodes. Optical image of the a) Nb$_2$CT$_X$ ASEI on Cu foil using the Langmuir–Blodgett scooping (LBS) method; b) Nb$_2$CT$_X$ Cu and Li-In foil (left), Nb$_2$CT$_X$ Li-In right after Nb$_2$CT$_X$ ASEI transfer to Li-In via roll pressing (middle) and Nb$_2$CT$_X$ Li-In after 3 days in a dry capsule (right). c) Large-area SEM image (left, top view), enlarged SEM image (middle, top view), and SEM image (right, cross section view) of the Nb$_2$CT$_X$ Li-In anode.
Fig. S9. XRD profile of the Nb$_2$CT$_x$ Li-In anode. The XRD peaks of Nb$_2$CT$_x$ and Li$_{13}$In$_3$ are indicated by the filled star and empty triangle symbols, respectively.
Fig. S10. Practical areal capacity measurement for Nb$_2$CT$_X$ Li-In and Li-In alloy anode using symmetric cell. Voltage profile taking at a fixed current density of 1 mA cm$^{-2}$ with voltage cut-off at 1V. The areal masses of Nb$_2$CT$_X$ Li-In and Li-In alloy anodes are 13.52 mg cm$^{-2}$ and 13.24 mg cm$^{-2}$. Capacities of Nb$_2$CT$_X$ Li-In and Li-In alloy anodes are 39.63 mAh cm$^{-2}$ (2926.64 mAh g$^{-1}$) and 39.97 mAh cm$^{-2}$ (3021.44 mAh g$^{-1}$). The areal capacity of Nb$_2$CT$_X$ Li-In alloy anode was slightly reduced due to pressure during roll pressing process for transfer of Nb$_2$CT$_X$ ASEI. The inset shows the enlarged voltage profile from 0 to 1.0 mAh cm$^{-2}$ with voltage profile of pure Li from Fig. 1b.
Fig S11. SEM surface morphologies of galvanostatic Li re-electrodeposition onto the f) Li anode and g) Li-In alloy anode after Li stripping of 4mAh cm$^{-2}$ at a fixed current density of 1 mAh cm$^{-2}$ and capacity of 4 mAh cm$^{-2}$.

Fig. S12. Galvanostatic Li stripping/plating voltage profiles for the Li (black), Li-In (red) and Nb$_2$CT$_x$ ASEI Li-In (blue) anodes. Voltage profiles were measured by Li|Li, Li-In|Li-In, and Li-In Nb$_2$CT$_x$|Nb$_2$CT$_x$ Li-In symmetric cells at a) 1 mA cm$^{-2}$/1 mAh cm$^{-2}$ and b) 2 mA cm$^{-2}$/2 mAh cm$^{-2}$. 
Fig. S13. a) Electrochemical full cell cycling performance with Li Nb$_2$CT$_x$|NCM811 based on discharge capacity (filled symbols, left axis) and Coulombic efficiency (empty symbols, right axis). The full cell was fabricated at an n/p ratio of 11.0 and by using an electrolyte of 8.6 µL mAh$^{-1}$. Operating conditions of full-cell cycling were 0.5 C for charging, 1.0 C for discharging, and voltage window of 2.7–4.3 V. The first highest discharge capacity point was measured at formation cycle (0.1 C charge & discharge); the yellow dashed line indicates 80% of the initial discharge capacity of each cell. b) Corresponding voltage profiles of Li Nb$_2$CT$_x$|NCM811.
Fig. S14. Nyquist plots after 1\textsuperscript{st}, 5\textsuperscript{th}, 10\textsuperscript{th}, and 15\textsuperscript{th} cycle for a) Li|NCM811, b) Li-In|NCM811, c) Li-In Nb\textsubscript{2}CT\textsubscript{X}|NCM811 full cell.

Fig. S15. Li-In foil thickness and roll-pressing results. a) Pristine Li-In foil from the company (Honzo metal, Japan), b) roll-pressed Li-In foil (average thickness of ~156 μm), and c) roll-pressed Li-In foil (average thickness of ~137 μm).
Table S1. The calculation of termination group formation energy and Li binding energy on the Nb$_2$C (MXene) surface. Images in the table are Nb$_2$CT$_X$ atomic structure with termination groups and Li adatoms. Cyan blue, brown, green, purple, red, and pink spheres are Niobium, Carbon, Lithium, Fluorine, Oxygen, and Hydrogen atom, respectively.
Table S2. Li-In foil thickness before and after roll-pressing from Fig. S15

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References