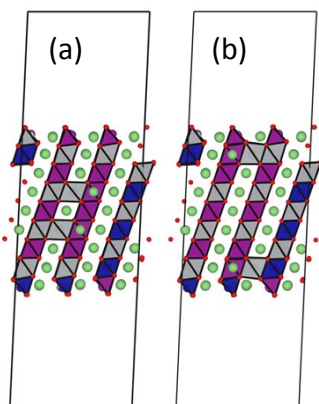




SUPPLEMENTARY INFORMATION

Figure S1. Slab polyhedral model for pristine $\text{LiNi}_{1/3}\text{Mn}_{1/3}\text{Co}_{1/3}\text{O}_2$ (NMC-111) surface (012) with cation disorder (a) Li/Ni exchange in the bulk region, (b) Li/Ni exchange in the surface region. The NiO_6 octahedra are represented in grey, CoO_6 octahedra are represented in blue and MnO_6 octahedra are represented in purple. The Li ions are indicated by green spheres.

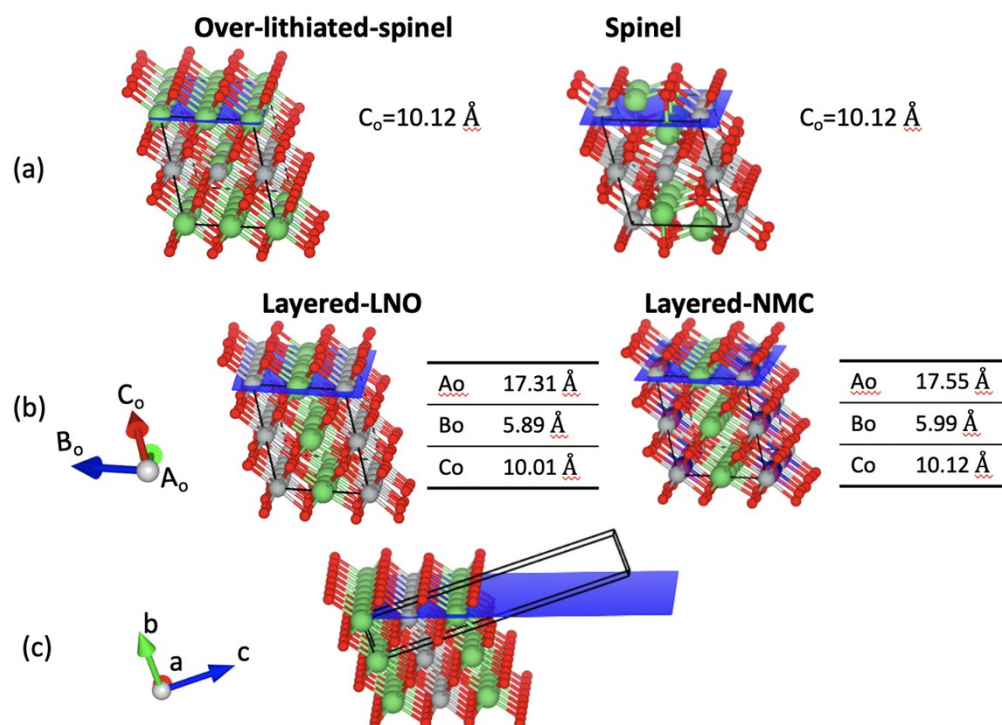


Figure S2. Schematic representation of the supercells used for modeling strain in the surface reconstruction layers. (a) Representation of the over-lithiated-spinel $\text{Li}_2\text{Ni}_2\text{O}_4$ and spinel LiNi_2O_4 . The value of C_0 for each cell is the initial value when the cell is fully relaxed. (b) Representation of the layered LNO and NMC supercells. The values of A_0 and B_0 (relaxed structures) are used as a reference to 'pin' the respective reconstructed layer under study. (c) layered

LiNiO₂ (012) surface ((012) plane shown in blue) in the R-3m original unit cell (black lines). Green spheres represent Li, grey spheres represent Ni, purple spheres represent Mn, blue spheres represent Co and red spheres represent O.

Estimation of configurational entropy:

Using the expression from reference [30]:

$$\Delta S = k_B \left[X_S \ln(X_S) + (1 - X_S) \ln(1 - X_S) - X_B \ln(X_B) - (1 - X_B) \ln(1 - X_B) \right]$$

Where:

ΔS : Configurational entropy [eV/K]

$X_S = 1/2$: surface composition (sites occupied)

$X_B = 1/6$: bulk composition (sites occupied)

$k_B = 8.617333\text{e-}5$: Boltzmann constant [eV/K]

Coordinates (VASP POSCAR format) of the slab shown in Figure S1a as a representative cell of the slab calculations:

```
Li Co Ni Mn O
1.0000000000000000
8.7666149138999998 0.0000000000000000 0.0000000000000000
-0.0037098171000000 15.3246712963000000 0.0000000000000000
4.4307786510999998 2.2311451227000001 50.1633890080000029
Li Co Ni Mn O
81 27 27 27 162
Direct
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0.5656884309999981 0.8870784040000004 0.5705810190000022
0.7315074799999977 0.3709734979999979 0.5722760559999998
0.0648404880000015 0.0376404900000011 0.5722760559999998
0.3981744649999968 0.0376404900000011 0.5722760559999998
0.7315074210000034 0.0376404900000011 0.5722760559999998
0.3981745239999981 0.7043074369999971 0.5722760559999998
0.0648405029999992 0.7043074369999971 0.5722760559999998
0.3981744949999992 0.3709734979999979 0.5722760559999998
0.0648404809999974 0.3709734979999979 0.5722760559999998

0.7315075989999968	0.7043074369999971	0.5722760559999998
0.3751962187532566	0.9379997354073941	0.6158133178808072
0.7230269952937816	0.5953174882675156	0.6186867286234539
0.0278600253306622	0.5948461688098985	0.6179328986329565
0.7163670408410833	0.2644618619433081	0.6159332543981815
0.0314326600653411	0.2634178716519254	0.6167359837467035
0.3864242635015175	0.2708756626276757	0.6184710590884733
0.6948771358690180	0.9319503534308882	0.6176518361853083
0.0441044266932181	0.9363436986612892	0.6198132370855161
0.3715293709651506	0.6026081461035351	0.6212168766987970
0.8730035878523918	0.0872593905677518	0.6230983095273084
0.5458846639784072	0.0871147265925299	0.6179737346475433
0.2170139656232293	0.7581045596194067	0.6220232328440737
0.5303161588283853	0.7571781530407806	0.6226713764315326
0.1948610471855680	0.0912131456252386	0.6214856602405570
0.5310358905399749	0.4246778928924191	0.6204919772781777
0.2197327969271733	0.4235970011086619	0.6207914514273929
0.8771865774401401	0.4231038252337068	0.6182054400440712
0.8741889683762305	0.7543136487575766	0.6180926696993668
0.5117233190633971	0.3087682839299610	0.6635221264297408
0.8572412759795945	0.9752946404233098	0.6639706504608490
0.5116085156728440	0.6412842592895543	0.6647746468692827
0.1789282825348852	0.9750078770607963	0.6638009691416463
0.1938396787839148	0.6422238265430137	0.6654582895150832
0.1952756516632823	0.3092600721623098	0.6633892402439224
0.5262514226830085	0.9760676495302548	0.6639559961140075
0.8509727860125503	0.6377133333675822	0.6661960911662796
0.8556577139365317	0.3091392741066930	0.6668583285037115
0.6835487997818337	0.1336387941791181	0.6715587724672712
0.6838641303562808	0.8015968044845310	0.6737382695478759
0.6904843353729131	0.4721434610726589	0.6731195218494045
0.0115370436797688	0.4708203672300947	0.6716654605153819
0.3491477379957608	0.4623488161710285	0.6695059263627431
0.0149531315905624	0.8017678165344977	0.6745652337242064
0.3474823712544213	0.7965122995308518	0.6714012821825186
0.3458682210677383	0.1346433596481575	0.6735982015935132
0.0164045127948856	0.1286648437879573	0.6701792779657874
0.6624896377184170	0.6846363945696660	0.7116355317216936
0.6654116391637177	0.3517256696859136	0.7097884471735445
0.3283805637027222	0.6923035210712091	0.7112657493000526
0.3294305652567047	0.3619740101328727	0.7095522261568893
0.9935526632778083	0.0260453063803098	0.7105044677606571
0.0025711050494998	0.6838814000299797	0.7111649765497488
0.6717658581075084	0.0205301274206705	0.7103767076886567
0.0035862322703626	0.3548062097901387	0.7093787811049284
0.3274988951762526	0.0152745417956995	0.7104808378313946

Coordinates (VASP POSCAR format) of the bulk structure shown in Figure S2a as a representative cell of the bulk calculations:

Overlithiated spinel pinned to Layered N

1.00000000000000

10.1201513481703209 0.0397464481402120 0.1610347742351373
15.2144956487688710 8.7437990750395187 0.2829411009856746
3.4497546352500890 0.0357566493173069 4.8965119951237899

Li Ni O

12 12 24

Direct

-0.0000972638932054 0.0000616161123552 -0.0000152920905915
0.0001408259927209 0.3332375973822944 0.9999376884151443
-0.0000285521652018 0.6666956507957637 0.0000414140425894
-0.0000102959430617 0.0000095801838182 0.4999866683065721
-0.0000109765269511 0.1666754303675504 0.5000009701777577
-0.0000326401289550 0.3333478043376684 0.5000622314489945
0.0000843841273868 0.4999664319045358 0.4999653814347114
0.0000315016953347 0.6666541880391240 0.4999467736259954
-0.0000653313417622 0.8333541225381675 0.5000440862909591
0.5000023199558059 0.1666597953653929 0.4999946283707469
0.4999679369790412 0.5000015324610515 0.5000360680341066
0.4999955029442203 0.8333554927088757 0.4999792409398615
0.0000656943304017 0.1666285796679125 -0.0000097993180561
0.0000099035012116 0.5000104200362403 -0.0000239877994058
0.0000034704632461 0.8333287668381969 -0.0000122594492966
0.4999860145901204 0.0000011241465582 0.0000306587312738
0.5000013668152368 0.1666645426446740 0.0000467860170842
0.5000214469686201 0.3333180897226272 -0.0000376321254255
0.4999288795630399 0.5000453516621869 -0.0001755236012898
0.4999193426936676 0.6667227972904686 -0.0001159183399177
0.4999747518416022 0.8333672632972025 -0.0000155277714906
0.4999876661122962 0.0000044453690578 0.5000103909671408
0.5000093152234415 0.3333148063195652 0.5000301463922253
0.4999570735174272 0.6666838576274012 0.5000527570681726
0.2663287367334638 0.9898574866045734 0.2226753213017746
0.2662948536820885 0.3231598909065558 0.2227545050664702
0.2662142260761317 0.6565318607464496 0.2228127560069130
0.2738405980690543 0.1499170908916931 0.2417936415804447
0.2737713573189623 0.4832931961497060 0.2418331613596749
0.2738007989814363 0.8166243044302142 0.2419185855324170
0.7315773524793090 0.0088873813633910 0.2285189313643340
0.7252707362319815 0.1863638246791091 0.2362573514483466
0.7317012795994977 0.3421804098744926 0.2285803802974900
0.7252386263025400 0.5197103666055617 0.2363103939366045
0.7315891750371328 0.6755747718842815 0.2285490395400605
0.7252525611030579 0.8530720500081206 0.2361573033507507
0.2684354087000146 0.9910976922823083 0.7715013628347759
0.2747291654212750 0.1469628244302903 0.7638034616517502
0.2683703695939698 0.3244385180599779 0.7714405469287158
0.2748045494970185 0.4802620359845237 0.7637557458262938
0.2684048348982917 0.6577756340612467 0.7714812917348451
0.2747439684134204 0.8135967590281099 0.7638024538216305
0.7262620855285308 0.1833406158344666 0.7581797149118752
0.7262445241080290 0.5167001343963791 0.7580998556903796
0.7261004402581077 0.8501145332754317 0.7582060940462434
0.7336662784436465 0.0101412213497361 0.7773011879123400

0.7337762885176368 0.3434489398356428 0.7772828621665588
0.7337390876897119 0.6768390844990995 0.7772244619214435