Supporting Information

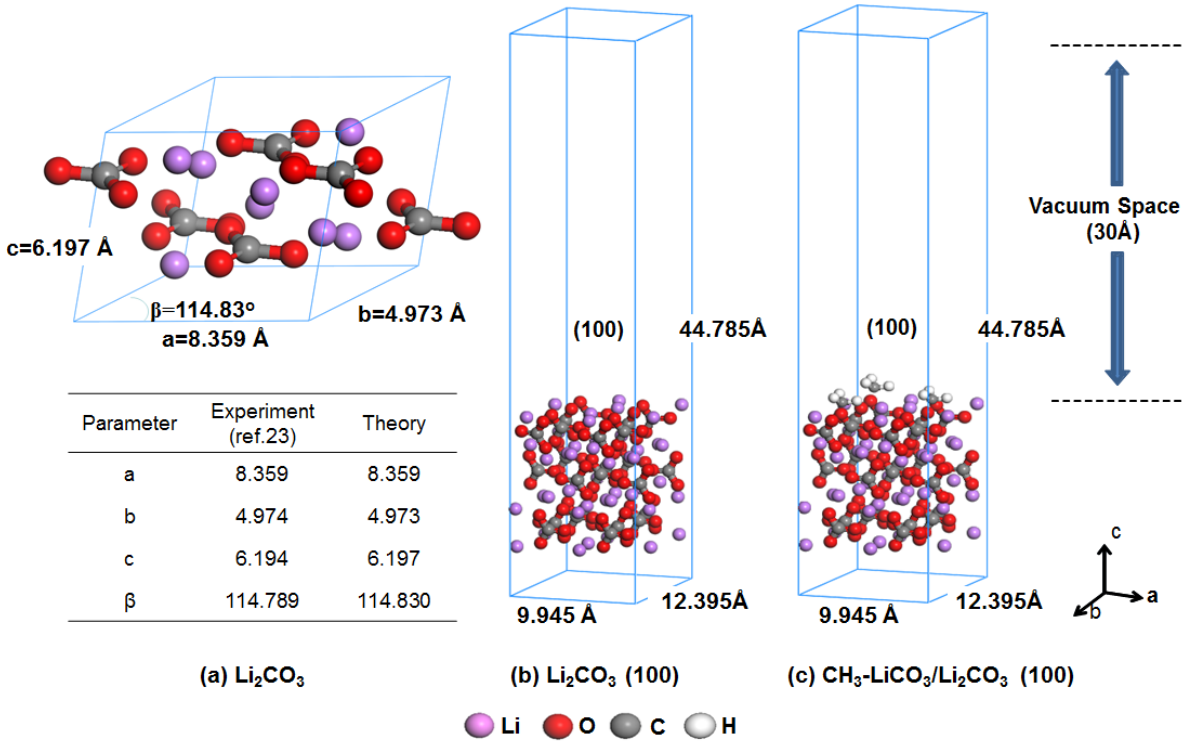
Self-terminated Artificial SEI Layer for Nickel-rich Layered Cathode Material via Mixed Gas Chemical Vapor Deposition

In Hyuk Son,*[,†,#] Jong Hwan Park,[†,#] Soonchul Kwon,[†] Junyoung Mun,[‡] and Jang Wook Choi*[,§]

[†]Energy Material Lab, Material Research Center, Samsung Advanced Institute of Technology, Samsung Electronics Co., LTD, 130 Samsung-ro, Yeongtong-gu, Suwon-si, Gyeonggi-do 443-803, Republic of Korea.

[‡]Department of Energy and Chemical Engineering, Incheon National University, 12-1, Songdo-dong, Yeonsu-gu, Incheon 406-840, Republic of Korea.

[§] Graduate School of Energy, Environment, Water, and Sustainability (EEWS), Korea Advanced Institute of Science and Technology (KAIST), Daejeon 305-701, Republic of Korea.



Parameter	Experiment (ref.23)	Theory
a	8.359	8.359
b	4.974	4.973
c	6.194	6.197
β	114.789	114.830

Figure S1. (a) Unit cell structure of Li_2CO_3, and model structures of (b) 2x4 Li_2CO_3 (100) and (c) 2x4 CH_3-$LiCO_3$/Li_2CO_3 (100) for DFT calculation. Inset table shows the lattice parameters of Li_2CO_3 to evaluate the model structure in comparison with experiment results.

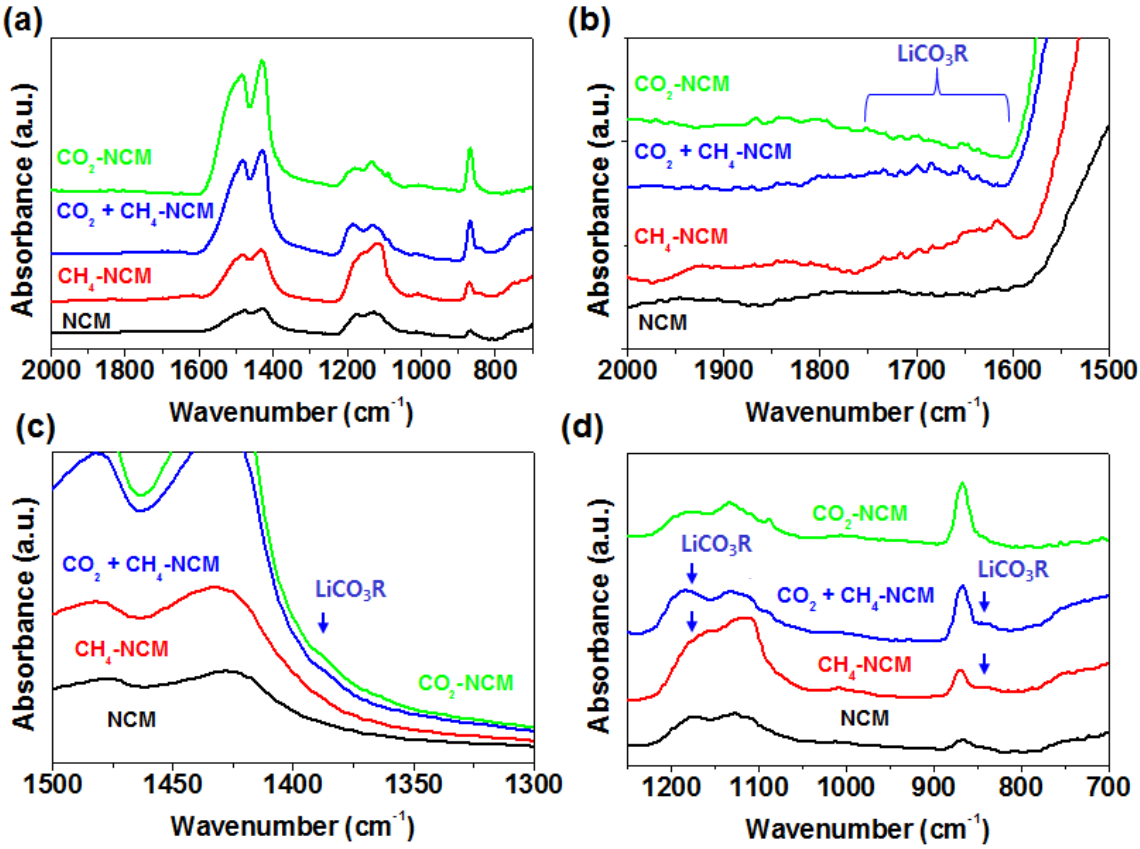


Figure S2. (a-d) *Ex-situ* ATR-FTIR spectra of pristine-NCM, CO_2-NCM, CH_4-NCM, and CO_2+CH_4-NCM after reaction for 30 min.

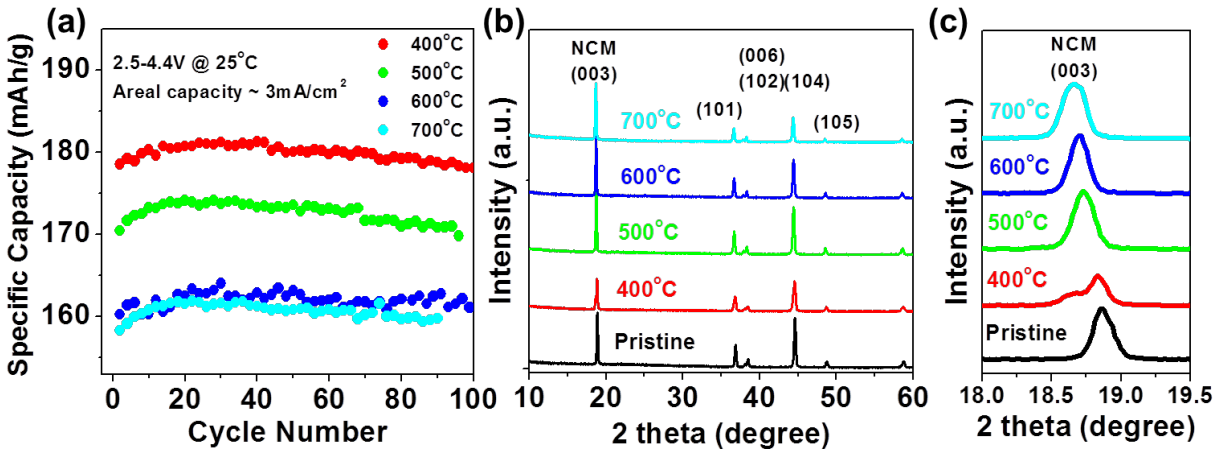


Figure S3. (a) The specific discharge capacity of CO_2+CH_4-NCM with various CVD temperatures (400-700 ^oC), and their XRD patterns (b-c).

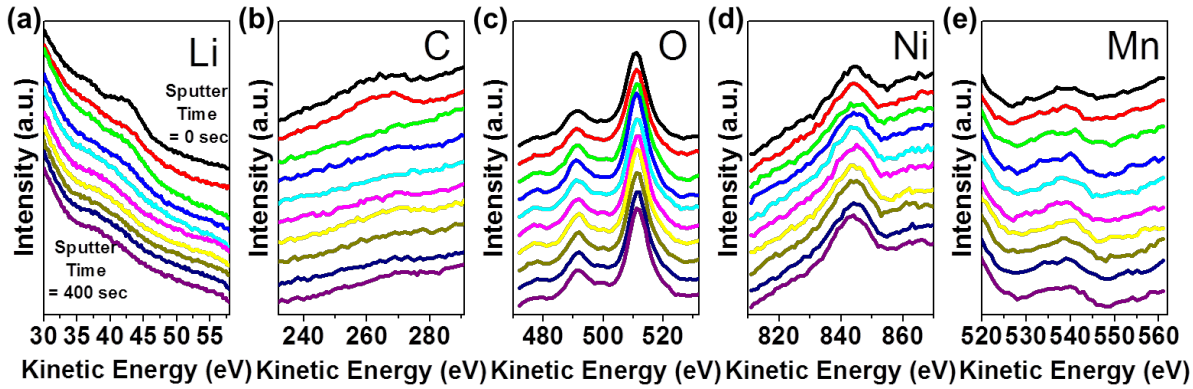


Figure S4. (a-e) Time-resolved Auger elemental depth profiles. Sputter time for each profile was 40 sec.

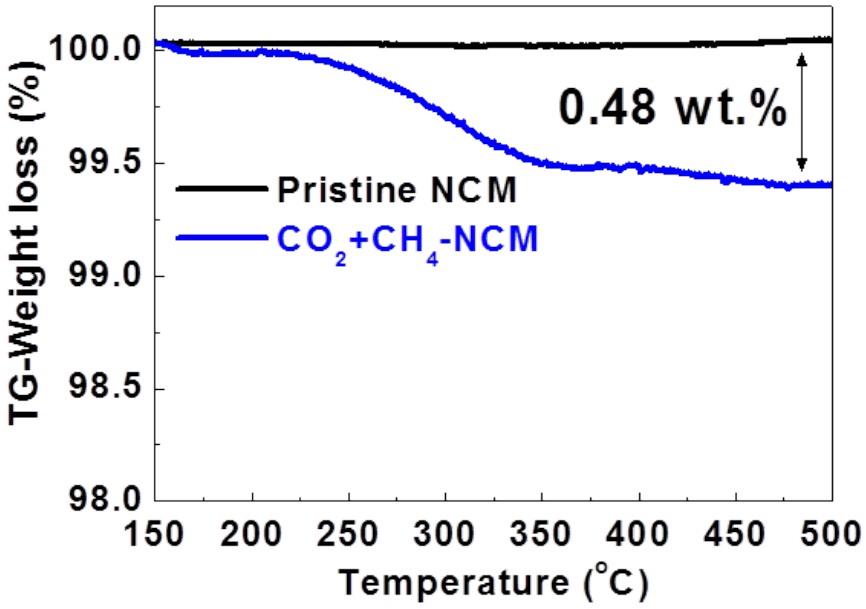


Figure S5. TGA profiles for the pristine and CO_2+CH_4-NCM.

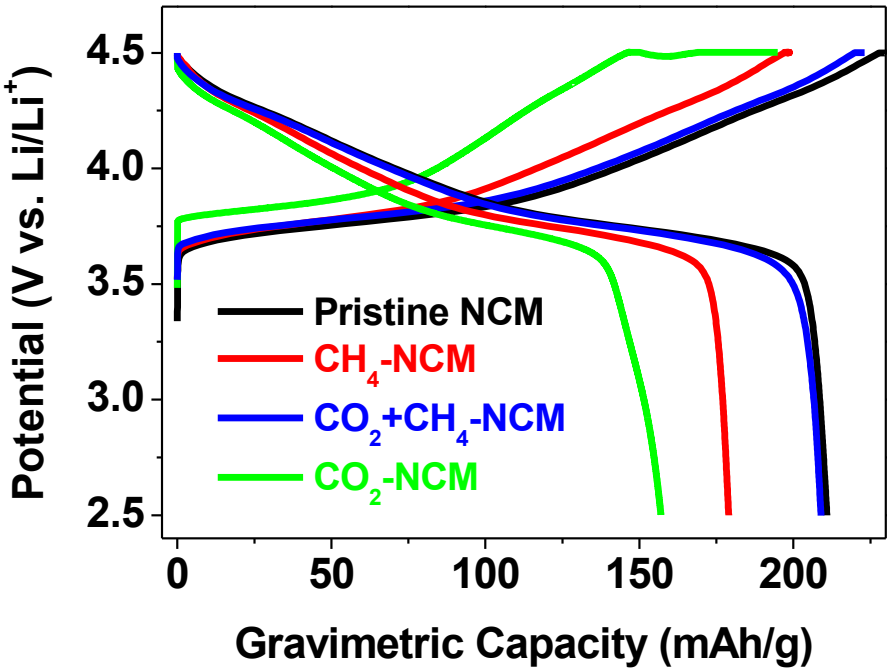


Figure S6. The initial (precycling) charge/discharge profiles of the prepared NCM half-cells at 2.5-4.5 V.

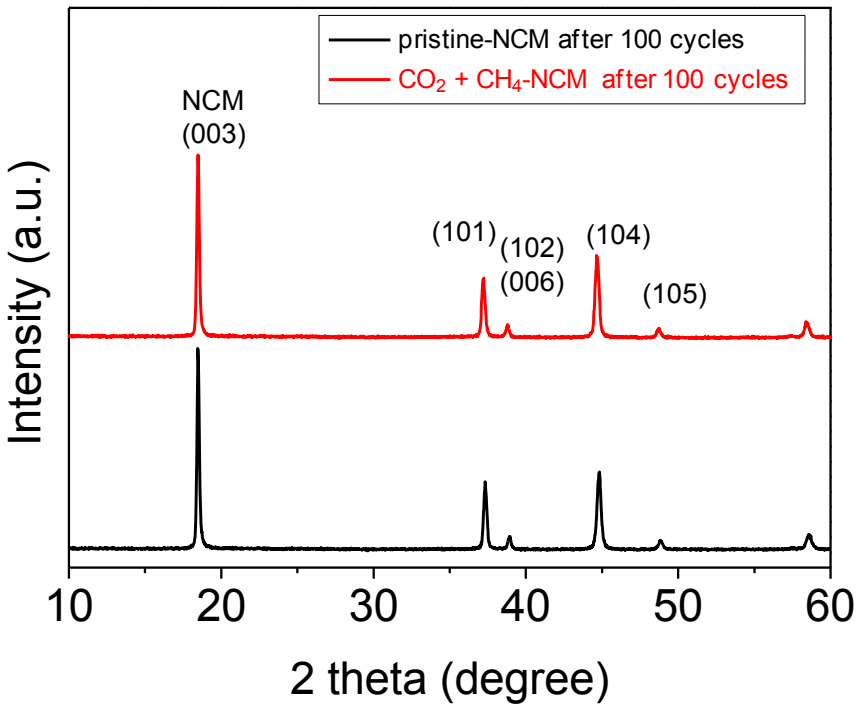


Figure S7. XRD profiles of the pristine and CO_2+CH_4 NCM after 100 cycles.

Sample	Time (h)	Extract concentration (mg/L)			
		Li	Mn	Co	Ni
Pristine-NCM	1	0.2	0.2	0.0	0.5
	3	0.4	0.6	0.2	1.2
	7	0.4	0.7	0.2	1.6
CO_2+CH_4-NCM	1	0.1	0.0	0.0	0.0
	3	0.2	0.0	0.0	0.0
	7	0.2	0.0	0.0	0.0

Table S1. Metal ion dissolution test for pristine and CO_2+CH_4 NCM using inductively coupled plasma-atomic emission spectroscopy (ICP-AES, ICP-AES_S, IPS-8100, Shimadzu). 0.1 g of the active material was dipped in 20 mL of the electrolyte co-solvents (EC:DEC:FEC=2:6:2=*v*:*v*:*v*) at 50 ^{o}C for 1, 3, and 7 hrs.