

Supporting Information

Investigations of ligand hole of Ni-rich layered cathodes: a new organic coverage to battery performance enhancement

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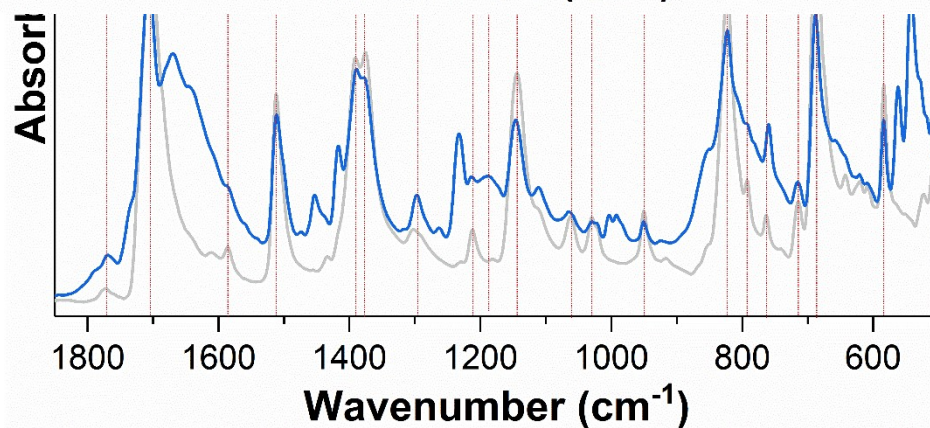
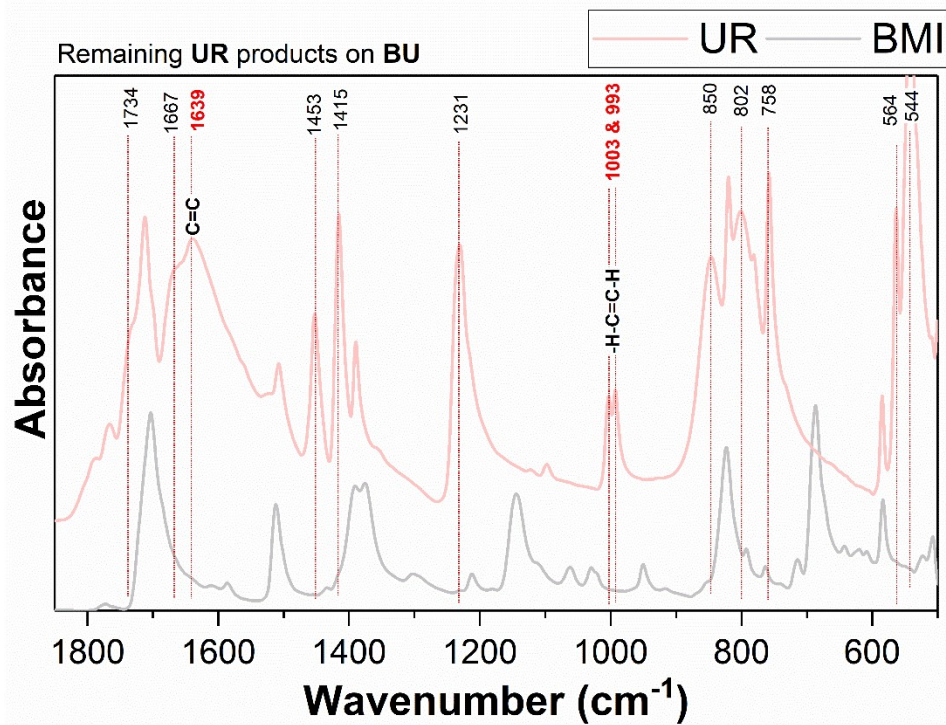
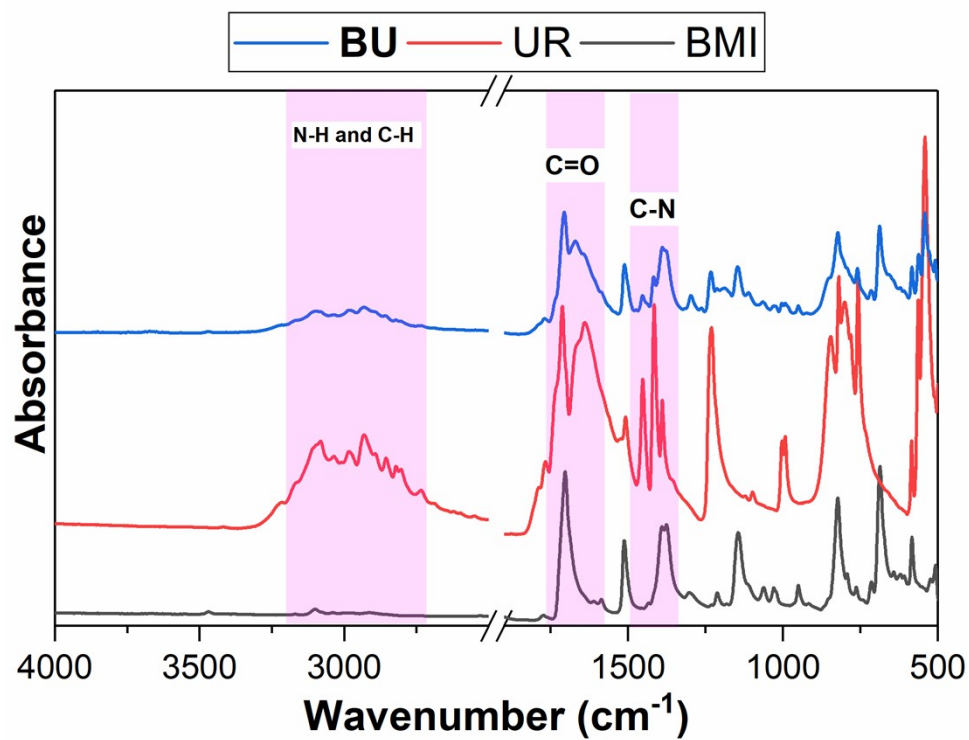


Figure S1 (a) The full ATR-FTIR spectra of BU, BMI and UR in the range of 400- 4000 cm^{-1} . (b) The ATR-FTIR spectra of remaining UR compound in BU structure. (c) The ATR-FTIR spectra of remaining BMI compound in BU structure.

Bismaleimide structure consists of two phenyl ring and two maleimide groups conjugated by a methyl group. Phenyl ring groups are represented by the absorbance peak between 830 - 650 cm^{-1} , while the strong absorbance at 1704 cm^{-1} is correspond to C=O of maleimide group, which follows by the absorbance peak of 1587 cm^{-1} from its C=C bond.

Uracil is heterocyclic ring which contained two carbonyl group and two N-H groups which represented by the strong absorbance intensit at the peak area around $\sim 1700 \text{ cm}^{-1}$ and $\sim 3000 \text{ cm}^{-1}$, respectively. BU was fabricated from the mixture of bismaleimide (BMI) and uracil(UR) in NMP solvent under continous stirring in an oil bath at 130 $^{\circ}\text{C}$. After synthesis, the phenyl, C=O and N-H group are remained in BU spectrum (Figure b and c). Meanwhile, the C=C bond at 1587 cm^{-1} from BMI shows reduced in absorbance intensity following by the appearenace of C-C bond at 1189 cm^{-1} in BU structure. This could indicated that BMI underwent ring opening reaction through its C=C bond maleimide and transforming into C-C bond in BU structure. In addition, the C=C bond from heterocyclic UR was almost completely dissapear on BU structure, indicating that UR also underwent ring opening on its C=C and bonded with BMI.

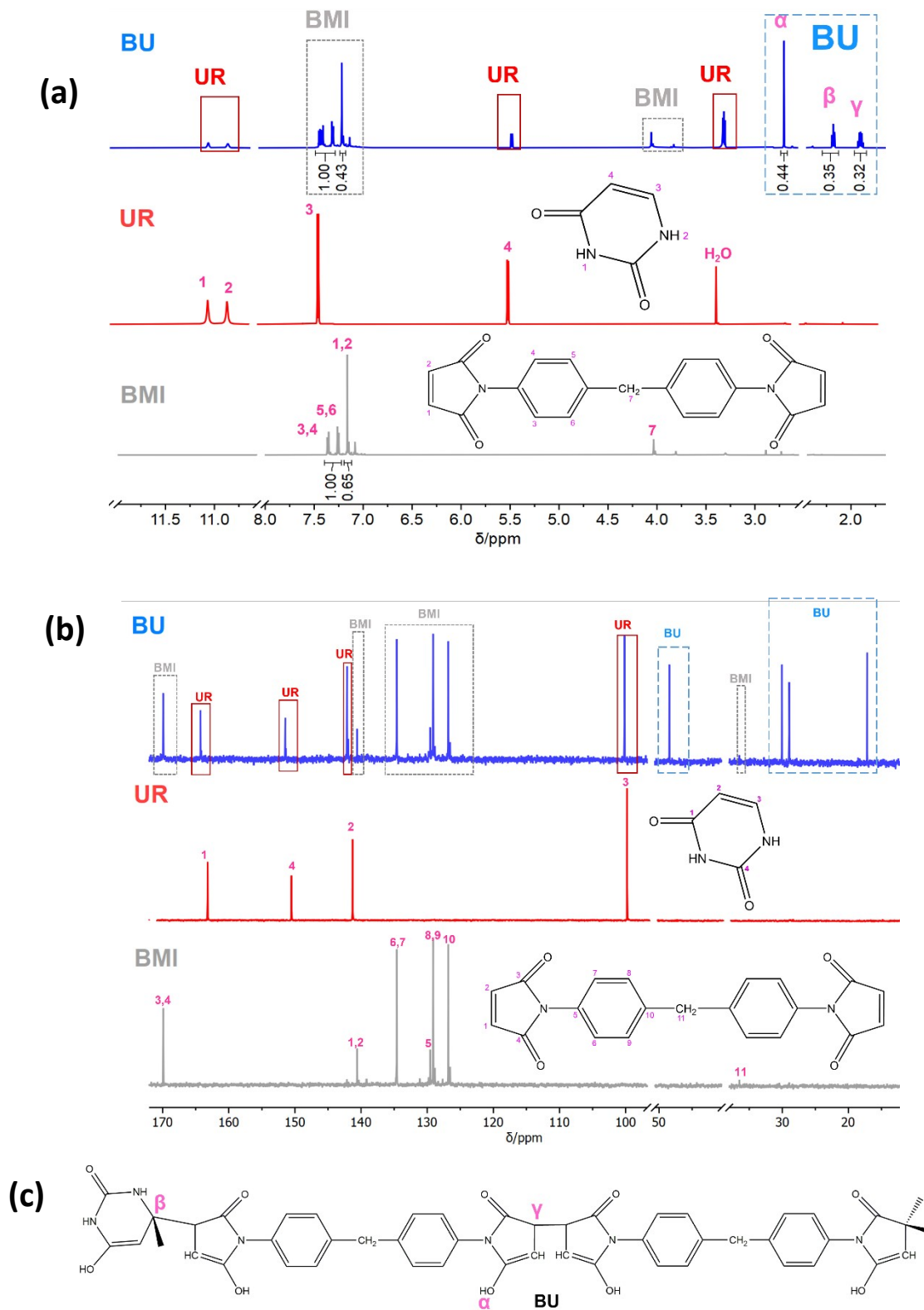


Figure S2 The ^1H and ^{13}C NMR spectra of BU, UR and BMI (a,b). The proposed structure of BU based on NMR and FTIR results (c).

The ^1H NMR spectra of BMI displayed the maleimide groups, denoted as “1” and “2” at δ (ppm) of 7.16. Meanwhile, the phenyl ring groups of BMI are denoted as “3”, “4”, “5”, and “6” which located at δ (ppm) of 7.35 and 7.26, respectively. The proton in methyl group that conjugated two phenyl groups in BMI is denoted as peak “7” at δ (ppm) of 4.03.

In the ^1H NMR spectra of Uracil (UR), proton that attached to nitrogen atom denoted as “1” and “2” at δ (ppm) of 10.99 and 10.79, respectively. The proton attached to carbon atom of $\text{C}=\text{C}$ displayed as peak “3” and “4” and located at δ (ppm) of 7.39 and 5.45, respectively.

After synthesis, both BMI and UR still retained its presence on BU structure. Three additional peaks appeared in BU structure, denoted as “ α ”, “ β ” and “ γ ”, correspond to the protons attached on C-OH and newly C-C bonds formed during synthesized (Figure c).

Table S1 showed the calculated integral area of maleimide to phenyl groups in BMI structure. The ratio of maleimide to phenyl ring displayed 0.6:1 ratio. However, the ratio of maleimide decreased to 0.4 in BU structure due to bond breaking of $\text{C}=\text{C}$ to bonded with another BMI or UR. Moreover the $\text{C}=\text{C}$ bond breaking also followed by the presence of C-OH with nearly equal ratio as the maleimide. This indicated that the formation of C-OH and remaining $\text{C}=\text{C}$ in maleimide are equal in BU structure. The presence of C-OH has been mentioned in the manuscript (page 4) as the result of catalytic conversion of $\text{C}=\text{O}$ into C-OH bonding via hydride anion transfer. Previous, the C-OH functional group also presence on another work using oligomer due to a proton donor from NMP solvent to carbonyl oxygen. In addition, ^{13}C NMR also spotted the presence of C-OH at δ (ppm) of 48.43.

Table S1 Integration area ratio obtained by ^1H -NMR for BMI and BU.

Sample	(Proton attached in Phenyl ring)	(Proton attached in maleimide)			
	7.37 and 7.26 ppm	7.16 ppm	2.69 ppm (α)	2.17 ppm (β)	1.89 ppm (γ)
BMI	1	0.602	-	-	-
BU	1	0.4	0.42	0.31	0.3

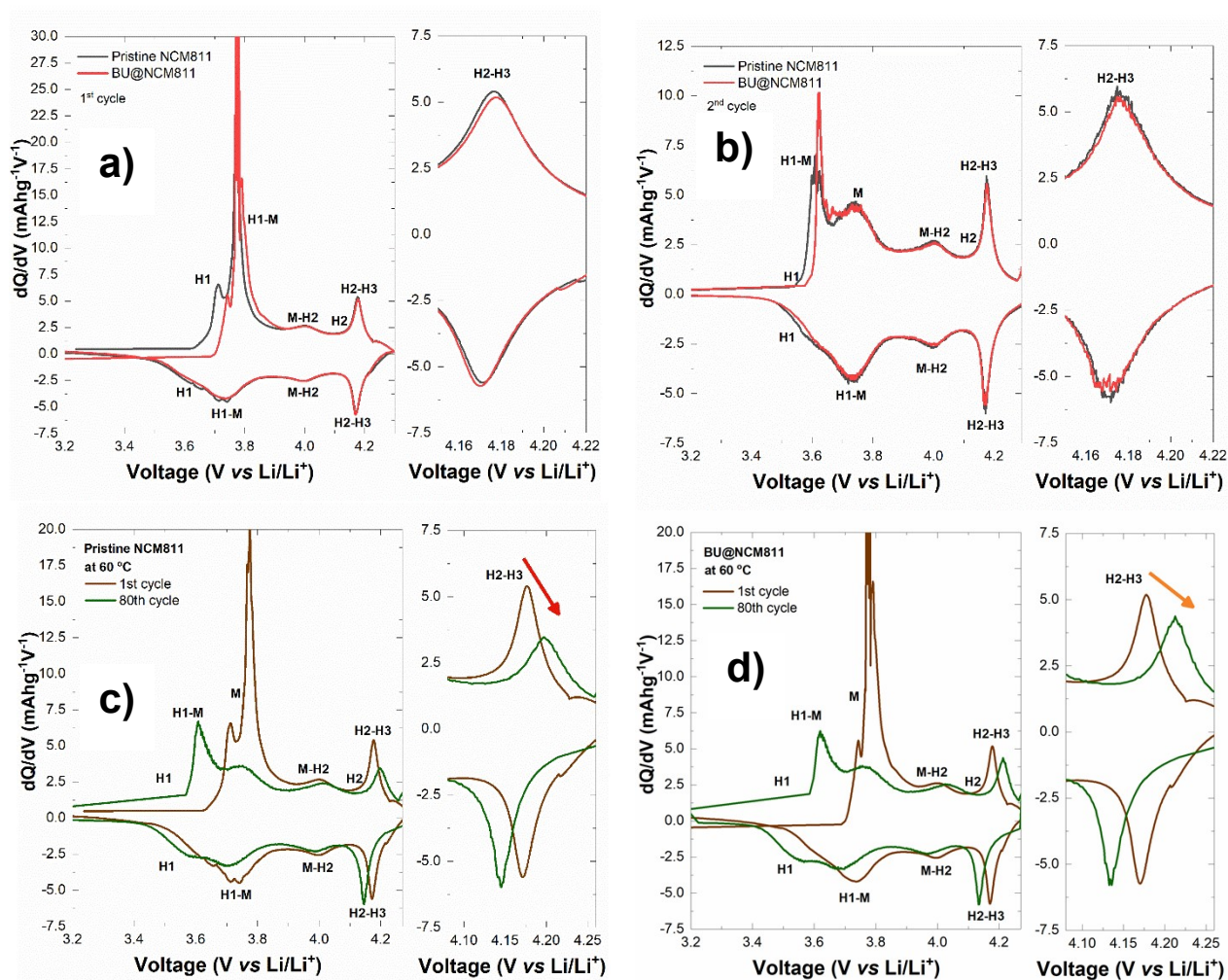


Figure S3 Differential capacity (dQ/dV) plots of pristine NMC811 and OC@NMC811 in (a) the first cycle and (b) the second cycle. Differential capacity (dQ/dV) plots of (c) pristine NMC811 and (d) OC@NMC811 at the 2nd and the 80th cycles. The enlarged section on the right highlight the H2-H3 region for detailed comparison.

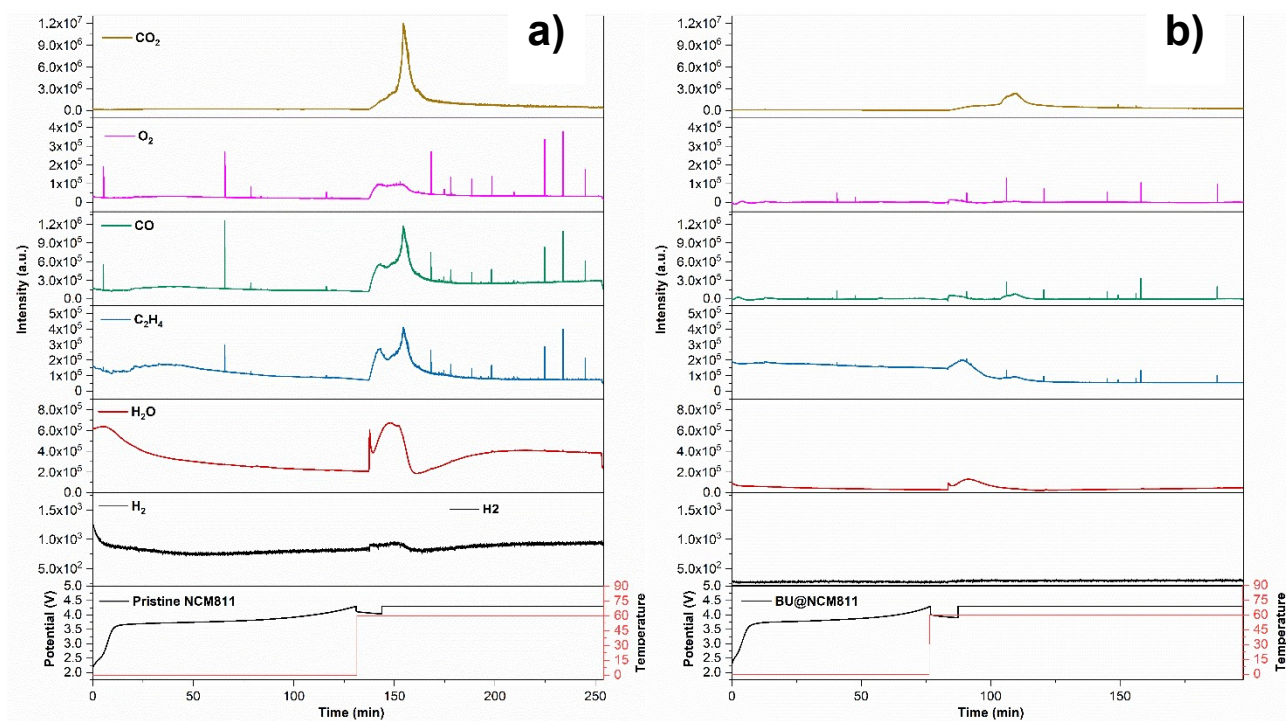


Figure S4 *Operando* GCMS analysis of two electrodes in the first cycle at room temperature.

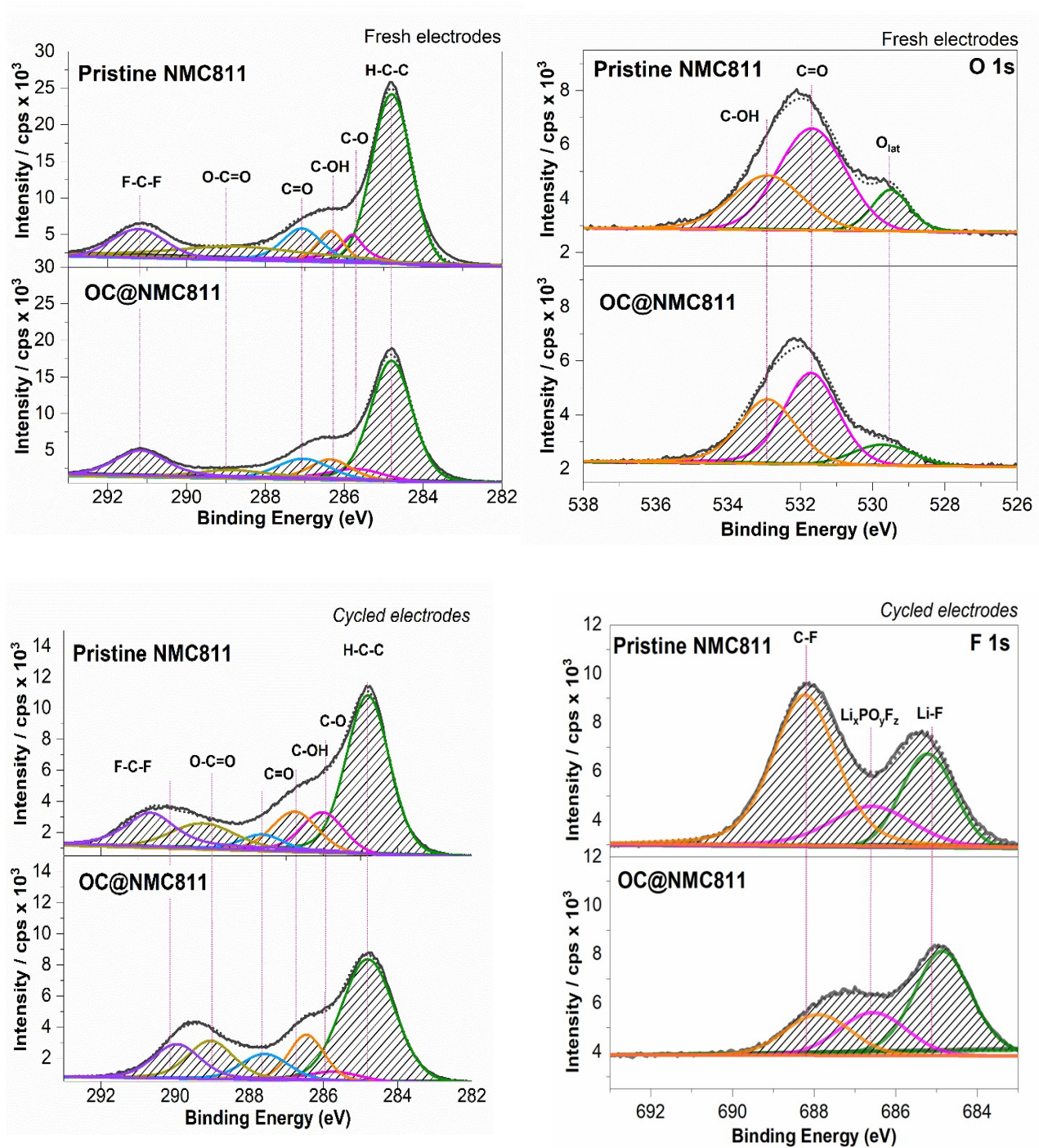


Figure S5 XPS analysis (a) C1s and (b) O1s spectra of two electrodes in fresh state. XPS analysis (c) C1s and (d) F1s spectra of two electrodes in cycled state.