

# Supplementary Information

## Mechanisms underlying the leaching process for **LiNi<sub>0.6</sub>Co<sub>0.2</sub>Mn<sub>0.2</sub>O<sub>2</sub> with and without H<sub>2</sub>O<sub>2</sub>**

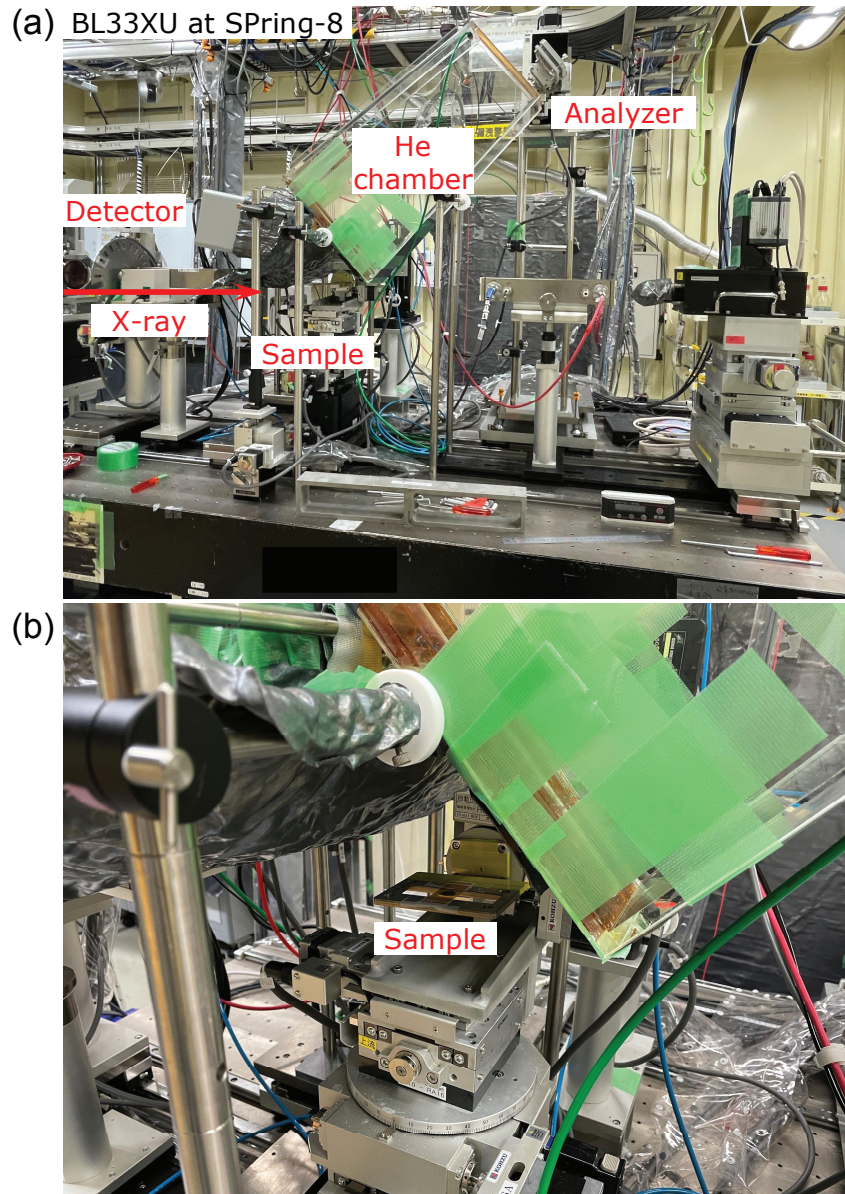
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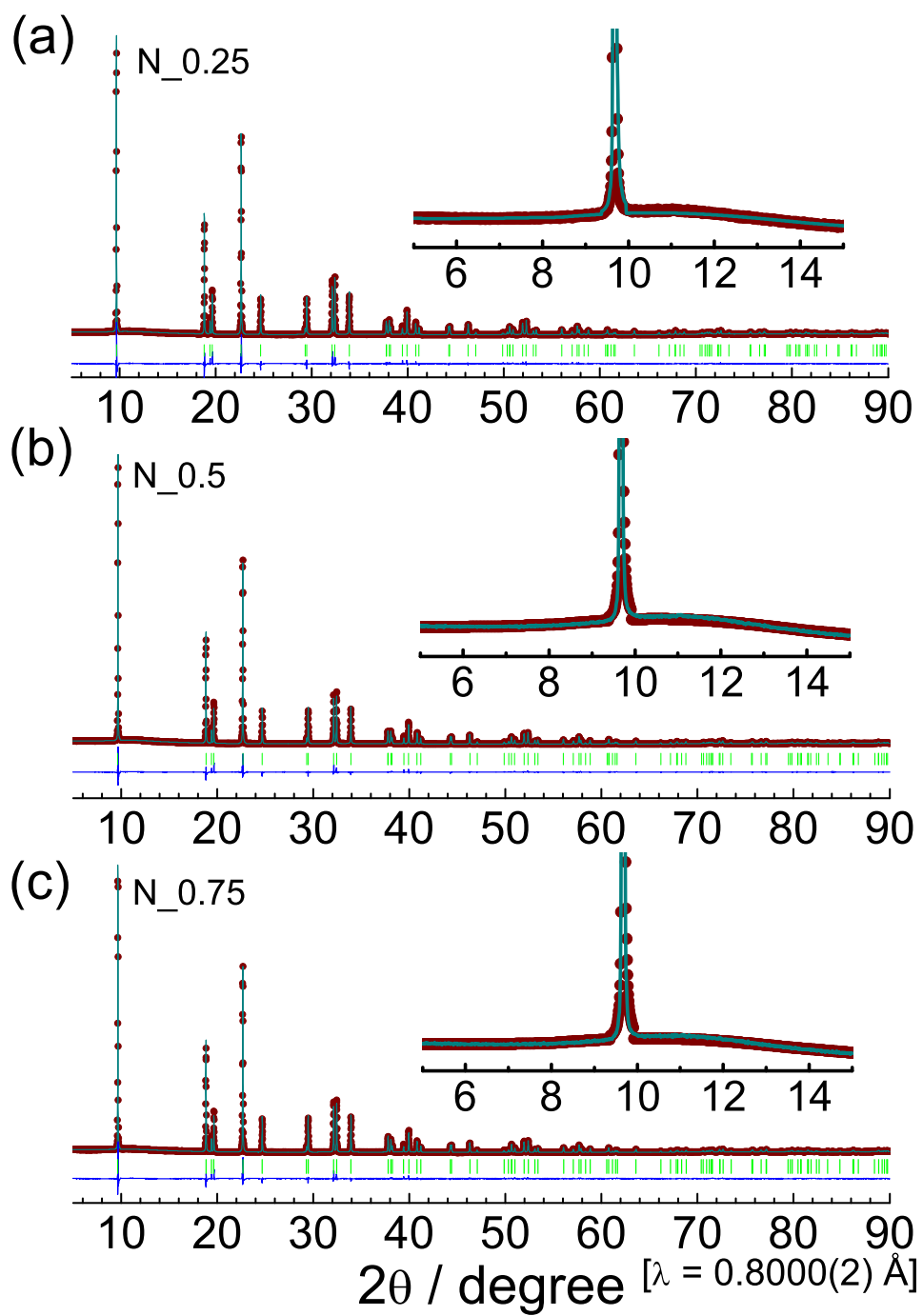
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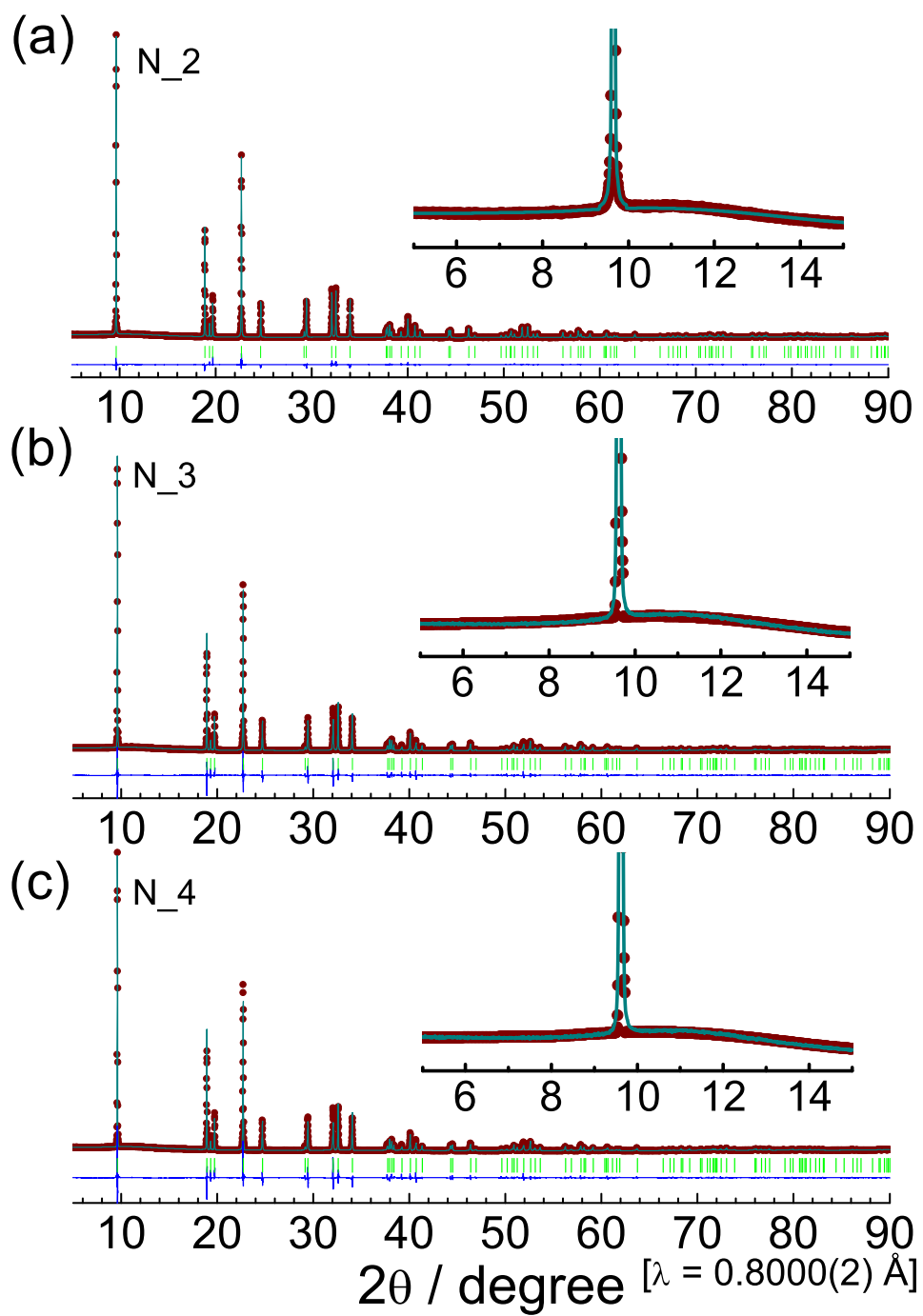


**Fig. S1:** Setup for the XRS measurements: (a) overall and (b) enlarged photograph of the sample holder.





**Fig. S2:** Rietveld analysis results after HNO<sub>3</sub> leaching without H<sub>2</sub>O<sub>2</sub>: (a) N<sub>0.25</sub>, (b) N<sub>0.5</sub>, and (c) N<sub>0.75</sub>.



**Fig. S3:** Rietveld analysis results after HNO<sub>3</sub> leaching without H<sub>2</sub>O<sub>2</sub>: (a) N<sub>2</sub>, (b) N<sub>3</sub>, and (c) N<sub>4</sub>.

**Table S1:** Structural parameters of N\_0.25, N\_0.5, N\_0.75, and N\_1.5 as determined by Rietveld analyses<sup>a</sup>

Sample	Phase	Atom	Wyckoff position	$g$	$x$	$y$	$z$	$B_{\text{iso}}$ ( $\text{\AA}^2$ )
N_0.25	NCM622	Li	$3b$	1.0	0	0	0.5	1.0
		Ni	$3a$	0.6	0	0	0	0.2(1)
		Co	$3a$	0.2	0	0	0	0.2(1)
		Mn	$3a$	0.2	0	0	0	0.2(1)
		O	$6c$	1.0	0	0	0.258(1)	0.4(1)
Space group: $R\bar{3}m$ , $a_{\text{h}} = 2.8658(1)$ $\text{\AA}$ , and $c_{\text{h}} = 14.229(1)$ $\text{\AA}$ $R_{\text{wp}} = 7.74$ % and $S = 0.93$ .								
N_0.5	NCM622	Li	$3b$	1.0	0	0	0.5	1.0
		Ni	$3a$	0.6	0	0	0	0.3(1)
		Co	$3a$	0.2	0	0	0	0.3(1)
		Mn	$3a$	0.2	0	0	0	0.3(1)
		O	$6c$	1.0	0	0	0.259(1)	0.5(1)
Space group: $R\bar{3}m$ , $a_{\text{h}} = 2.8646(1)$ $\text{\AA}$ , and $c_{\text{h}} = 14.229(1)$ $\text{\AA}$ $R_{\text{wp}} = 5.80$ % and $S = 0.71$ .								
N_0.75	NCM622	Li	$3b$	0.97	0	0	0.5	1.0
		Ni	$3a$	0.6	0	0	0	0.3(1)
		Co	$3a$	0.2	0	0	0	0.3(1)
		Mn	$3a$	0.2	0	0	0	0.3(1)
		O	$6c$	1.0	0	0	0.259(1)	0.5(1)
Space group: $R\bar{3}m$ , $a_{\text{h}} = 2.8639(1)$ $\text{\AA}$ , and $c_{\text{h}} = 14.236(1)$ $\text{\AA}$ $R_{\text{wp}} = 6.29$ % and $S = 0.77$ .								
N_1.5	NCM622	Li	$3b$	1.0	0	0	0.5	1.0
		Ni	$3a$	0.6	0	0	0	0.3(1)
		Co	$3a$	0.2	0	0	0	0.3(1)
		Mn	$3a$	0.2	0	0	0	0.3(1)
		O	$6c$	1.0	0	0	0.260(1)	0.5(1)
Space group: $R\bar{3}m$ , $a_{\text{h}} = 2.8582(1)$ $\text{\AA}$ , and $c_{\text{h}} = 14.269(1)$ $\text{\AA}$ $R_{\text{wp}} = 3.74$ % and $S = 0.45$ .								

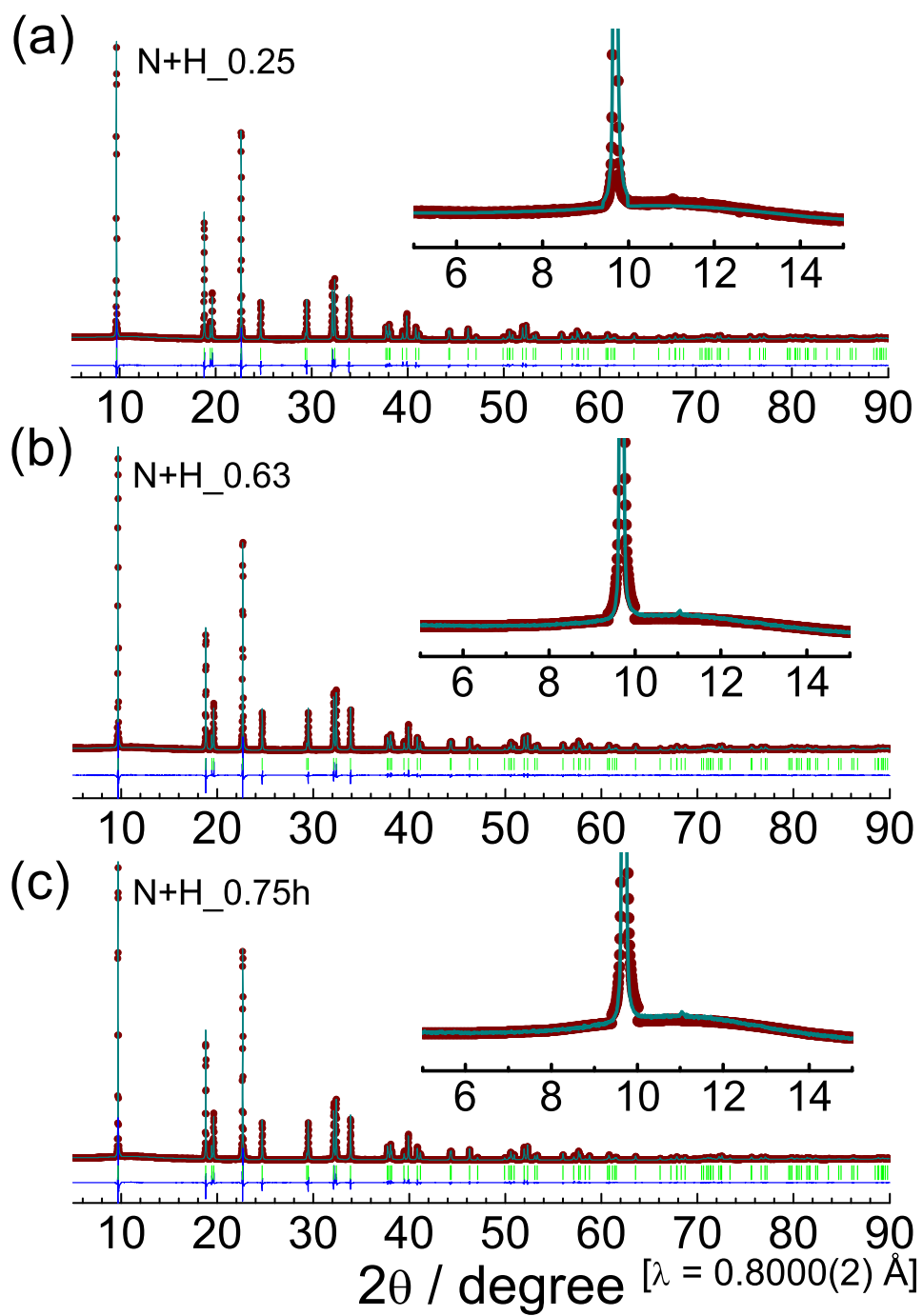
<sup>a</sup> $R_{\text{wp}}$  and  $S$  are the weighted reliability index and goodness-of-fit indicator, respectively, in the Rietveld analysis.

**Table S2:** Structural parameters of N\_2, N\_3, N\_4, and N\_8 as determined by Rietveld analyses<sup>a</sup>

Sample	Phase	Atom	Wyckoff position	$g$	$x$	$y$	$z$	$B_{\text{iso}}$ ( $\text{\AA}^2$ )
N_2	NCM622	Li	$3b$	1.0	0	0	0.5	1.0
		Ni	$3a$	0.6	0	0	0	0.2(1)
		Co	$3a$	0.2	0	0	0	0.2(1)
		Mn	$3a$	0.2	0	0	0	0.2(1)
		O	$6c$	1.0	0	0	0.260(1)	0.5(1)
Space group: $R\bar{3}m$ , $a_{\text{h}} = 2.8571(1)$ $\text{\AA}$ , and $c_{\text{h}} = 14.270(1)$ $\text{\AA}$ $R_{\text{wp}} = 4.00$ % and $S = 0.49$ .								
N_3	NCM622	Li	$3b$	0.97	0	0	0.5	1.0
		Ni	$3a$	0.6	0	0	0	0.2(1)
		Co	$3a$	0.2	0	0	0	0.2(1)
		Mn	$3a$	0.2	0	0	0	0.2(1)
		O	$6c$	1.0	0	0	0.261(1)	0.4(1)
Space group: $R\bar{3}m$ , $a_{\text{h}} = 2.8499(1)$ $\text{\AA}$ , and $c_{\text{h}} = 14.287(1)$ $\text{\AA}$ $R_{\text{wp}} = 10.6$ % and $S = 1.31$ .								
N_4	NCM622	Li	$3b$	0.97	0	0	0.5	1.0
		Ni	$3a$	0.6	0	0	0	0.3(1)
		Co	$3a$	0.2	0	0	0	0.3(1)
		Mn	$3a$	0.2	0	0	0	0.3(1)
		O	$6c$	1.0	0	0	0.259(1)	0.5(1)
Space group: $R\bar{3}m$ , $a_{\text{h}} = 2.8495(1)$ $\text{\AA}$ , and $c_{\text{h}} = 14.290(1)$ $\text{\AA}$ $R_{\text{wp}} = 8.15$ % and $S = 1.23$ .								
N_8	NCM622 (98.7 wt%)	Li	$3b$	0.26	0	0	0.5	1.0
		Ni	$3a$	0.6	0	0	0	0.4(1)
		Co	$3a$	0.19	0	0	0	0.4(1)
		Mn	$3a$	0.2	0	0	0	0.4(1)
		O	$6c$	1.0	0	0	0.265(1)	0.3(1)
	$\gamma$ -NiOOH (1.3 wt%)	Ni	$3a$	1	0	0	0	0.4(1)
		O	$6c$	1	0	0	0.370(4)	0.3(1)
Space group: $R\bar{3}m$ , $a_{\text{h}} = 2.8213(1)$ $\text{\AA}$ , and $c_{\text{h}} = 14.482(1)$ $\text{\AA}$ for NCM622 Space group: $R\bar{3}m$ , $a_{\text{h}} = 2.813(18)$ $\text{\AA}$ , and $c_{\text{h}} = 20.747(24)$ $\text{\AA}$ for $\gamma$ -NiOOH $R_{\text{wp}} = 4.96$ % and $S = 0.64$ .								

<sup>a</sup> $R_{\text{wp}}$  and  $S$  are the weighted reliability index and goodness-of-fit indicator, respectively, in the Rietveld analysis.



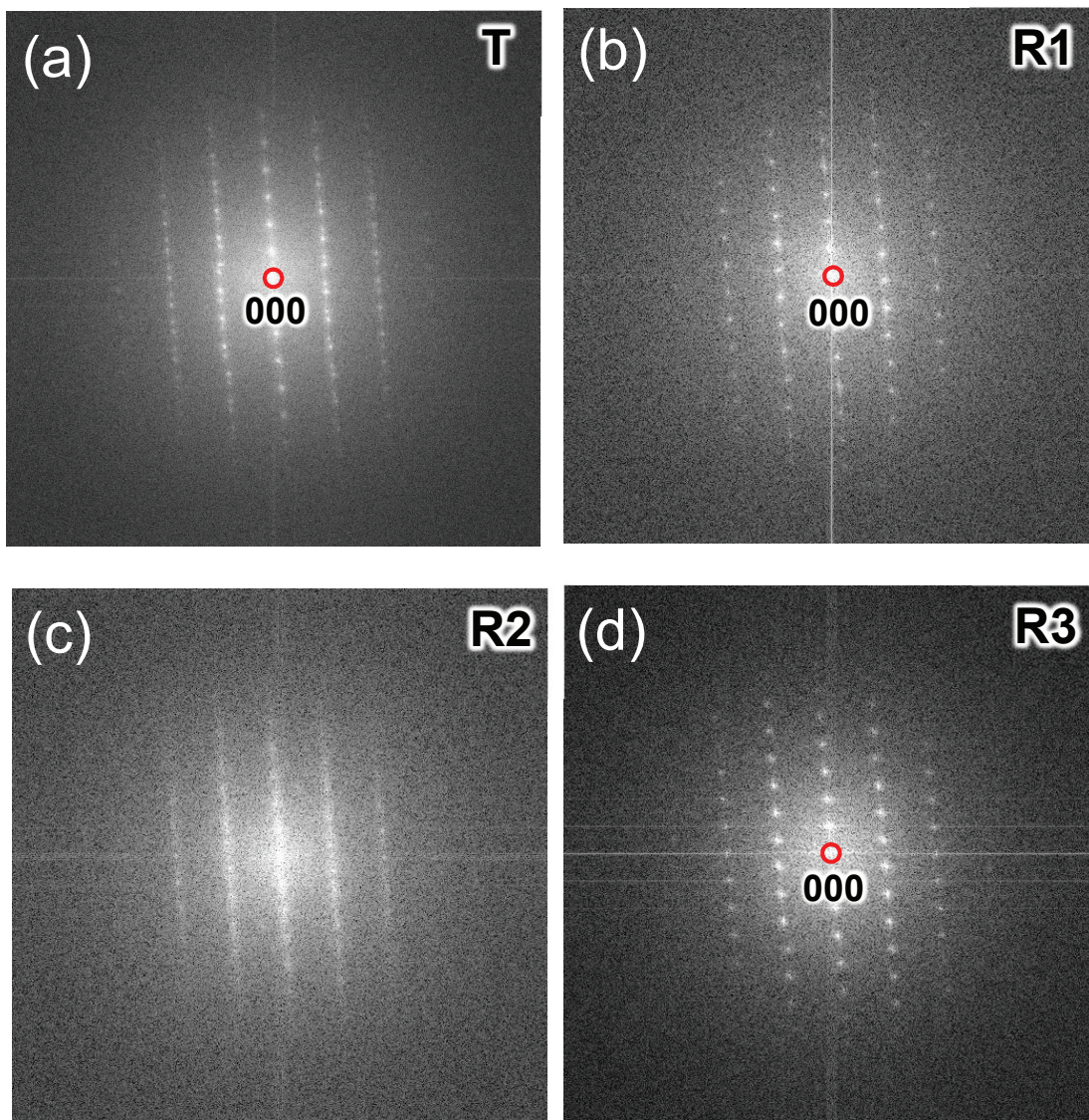


**Fig. S4:** Rietveld analysis results after  $\text{HNO}_3$  leaching with  $\text{H}_2\text{O}_2$ : (a) N+H.0.25, (b) N+H.0.63, and (c) N+H.0.75.

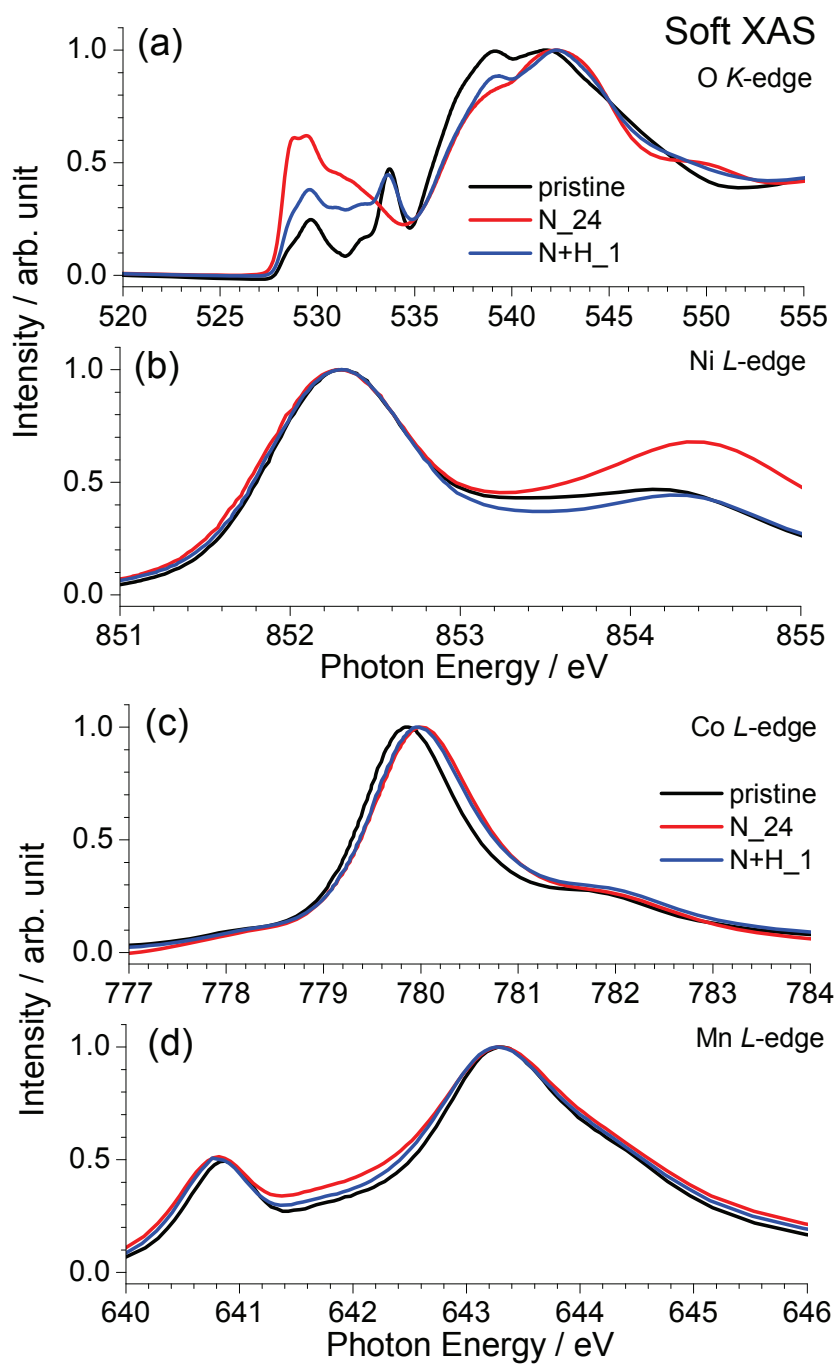
**Table S3:** Structural parameters of N+H.0.25, N+H.0.5, N+H.0.63, N+H.0.75, and N+H.2 as determined by Rietveld analyses<sup>a</sup>

Sample	Phase	Atom	Wyckoff position	$g$	$x$	$y$	$z$	$B_{\text{iso}}$ ( $\text{\AA}^2$ )
N+H.0.25	NCM622	Li	$3b$	1.0	0	0	0.5	1.0
		Ni	$3a$	0.6	0	0	0	0.2(1)
		Co	$3a$	0.2	0	0	0	0.2(1)
		Mn	$3a$	0.2	0	0	0	0.2(1)
		O	$6c$	1.0	0	0	0.259(1)	0.4(1)
Space group: $R\bar{3}m$ , $a_h = 2.8678(1)$ $\text{\AA}$ , and $c_h = 14.219(1)$ $\text{\AA}$ $R_{\text{wp}} = 9.07$ % and $S = 1.21$ .								
N+H.0.5	NCM622	Li	$3b$	1.0	0	0	0.5	1.0
		Ni	$3a$	0.6	0	0	0	0.3(1)
		Co	$3a$	0.2	0	0	0	0.3(1)
		Mn	$3a$	0.2	0	0	0	0.3(1)
		O	$6c$	1.0	0	0	0.259(1)	0.5(1)
Space group: $R\bar{3}m$ , $a_h = 2.8678(1)$ $\text{\AA}$ , and $c_h = 14.219(1)$ $\text{\AA}$ $R_{\text{wp}} = 8.09$ % and $S = 1.10$ .								
N+H.0.63	NCM622	Li	$3b$	0.97	0	0	0.5	1.0
		Ni	$3a$	0.6	0	0	0	0.3(1)
		Co	$3a$	0.2	0	0	0	0.3(1)
		Mn	$3a$	0.2	0	0	0	0.3(1)
		O	$6c$	1.0	0	0	0.259(1)	0.5(1)
Space group: $R\bar{3}m$ , $a_h = 2.8676(1)$ $\text{\AA}$ , and $c_h = 14.219(1)$ $\text{\AA}$ $R_{\text{wp}} = 8.77$ % and $S = 1.21$ .								
N+H.0.75	NCM622	Li	$3b$	1.0	0	0	0.5	1.0
		Ni	$3a$	0.6	0	0	0	0.3(1)
		Co	$3a$	0.2	0	0	0	0.3(1)
		Mn	$3a$	0.2	0	0	0	0.3(1)
		O	$6c$	1.0	0	0	0.259(1)	0.5(1)
Space group: $R\bar{3}m$ , $a_h = 2.8676(1)$ $\text{\AA}$ , and $c_h = 14.219(1)$ $\text{\AA}$ $R_{\text{wp}} = 8.98$ % and $S = 1.34$ .								
N+H.2	NCM622	Li	$3b$	1.0	0	0	0.5	1.0
		Ni	$3a$	0.6	0	0	0	0.4(1)
		Co	$3a$	0.2	0	0	0	0.4(1)
		Mn	$3a$	0.2	0	0	0	0.4(1)
		O	$6c$	1.0	0	0	0.260(1)	0.6(1)
Space group: $R\bar{3}m$ , $a_h = 2.8679(1)$ $\text{\AA}$ , and $c_h = 14.219(1)$ $\text{\AA}$ $R_{\text{wp}} = 5.76$ % and $S = 0.75$ .								

<sup>a</sup> $R_{\text{wp}}$  and  $S$  are the weighted reliability index and goodness-of-fit indicator, respectively, in the Rietveld analysis.

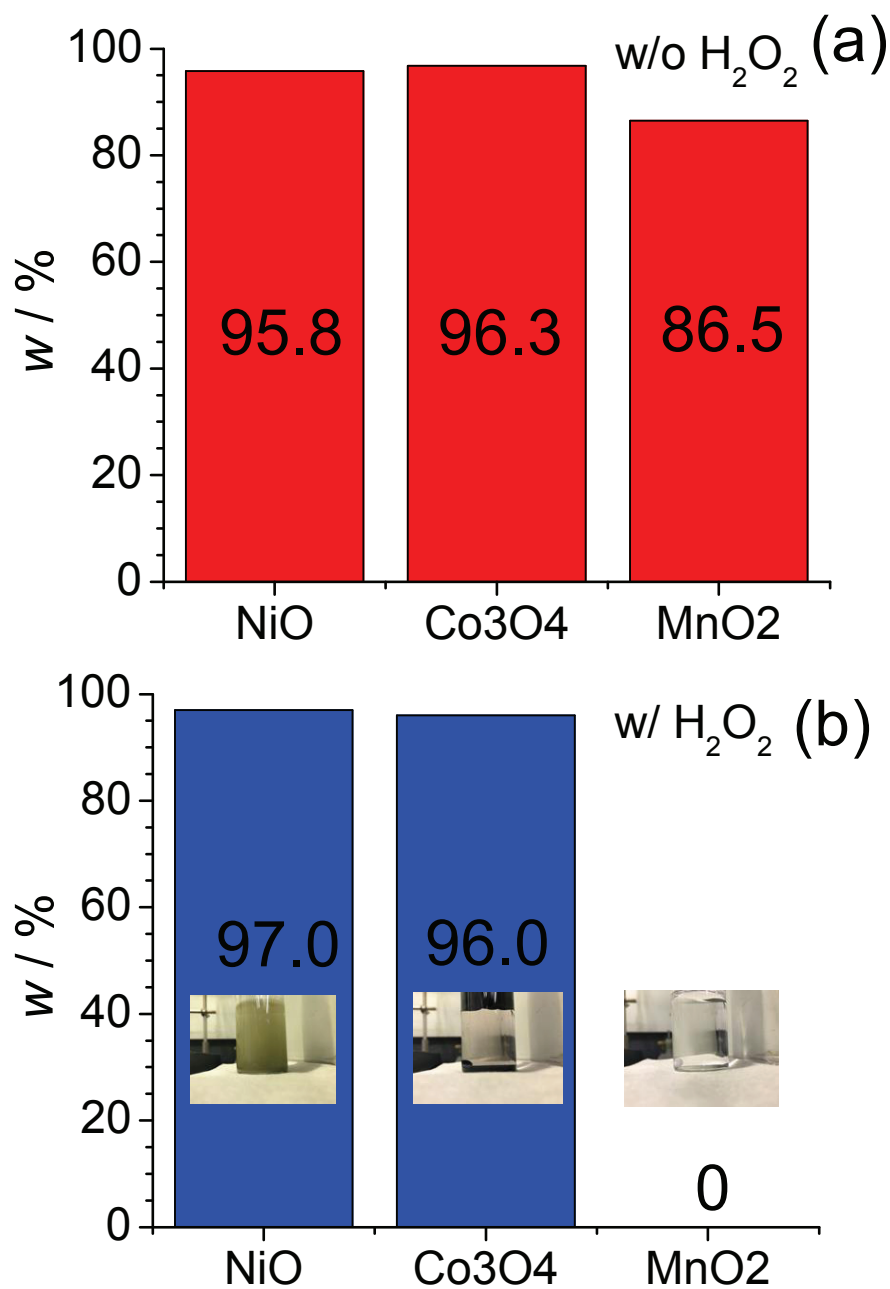


**Fig. S5:** Original FFT patterns obtained in the  $[100]$  or  $[110]$  direction from (a) the total area and from the regions labeled (c) R1, (d) R2, and (e) R3.



**Fig. S6:** Enlarged XANES spectra of pristine NCM622, N<sub>24</sub>, and N+H<sub>1</sub>: (a) O *K*-edge, (b) Ni *L*-edge, (c) Co *L*-edge, and (d) Mn *L*-edge.





**Fig. S7:** The weight fraction after 24 h of the HNO<sub>3</sub> leaching NiO, Co<sub>3</sub>O<sub>4</sub>, and MnO<sub>2</sub> (a) without H<sub>2</sub>O<sub>2</sub> and (b) with H<sub>2</sub>O<sub>2</sub>. The insets in (b) show photographs after leaching with H<sub>2</sub>O<sub>2</sub>.