Supplementary Materials

RBS and XAS Study of Lithium Nickel Cobalt Manganese Oxide/Lithium Lanthanum Zirconate/Zinc Ferrite Trilayer Structures Grown Using Radio Frequency Sputtering

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Section S1: Inductively coupled plasma mass spectrometry (ICP-MS) analysis

To further investigate the presence of lithium in the NLZ100, the ICP-MS technique is used. The sample (m) was prepared using 0.1452 gm of sample followed by digestion and dilution to a final volume (v) of 15 mL.

The ICP-MS yielded a lithium concentration (C_i) of 36.341 ppb for the undiluted sample with a relative standard deviation (RSD) of 0.8%. For the 10-fold diluted sample, a concentration of 4.383 ppb was obtained with an RSD of 3.6% (Table S4). Based on the sample mass and final digestion volume, the corresponding lithium concentration in NLZ100 was estimated using [47]

$$C = \frac{C_i \times v \times f}{m}$$

where C_i is the concentration, v is the volume of the diluted digestion solution, m is the mass of the sample used, and f is the dilution factor,

For undiluted sample, C_i = 36.341 ppb and f = 1

$$C_{diluted} = 3.76 \text{ mg/kg}$$

For diluted sample, C_i = 4.383 ppb and f = 10

$$C_{undiluted} = 4.53 \text{ mg/kg}$$

Therefore, the corresponding lithium concentration for diluted and undiluted samples was estimated to be 3.76-4.53 mg/kg.

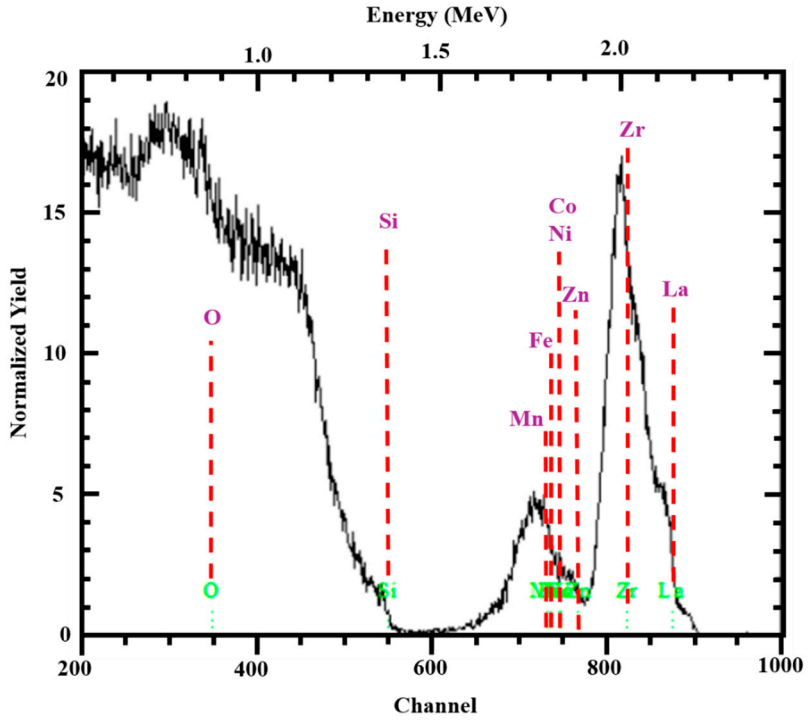


Figure S1: RBS spectra of NLZ25 showing the presence of all elements specific to ZFO, LLZO and NCM layers

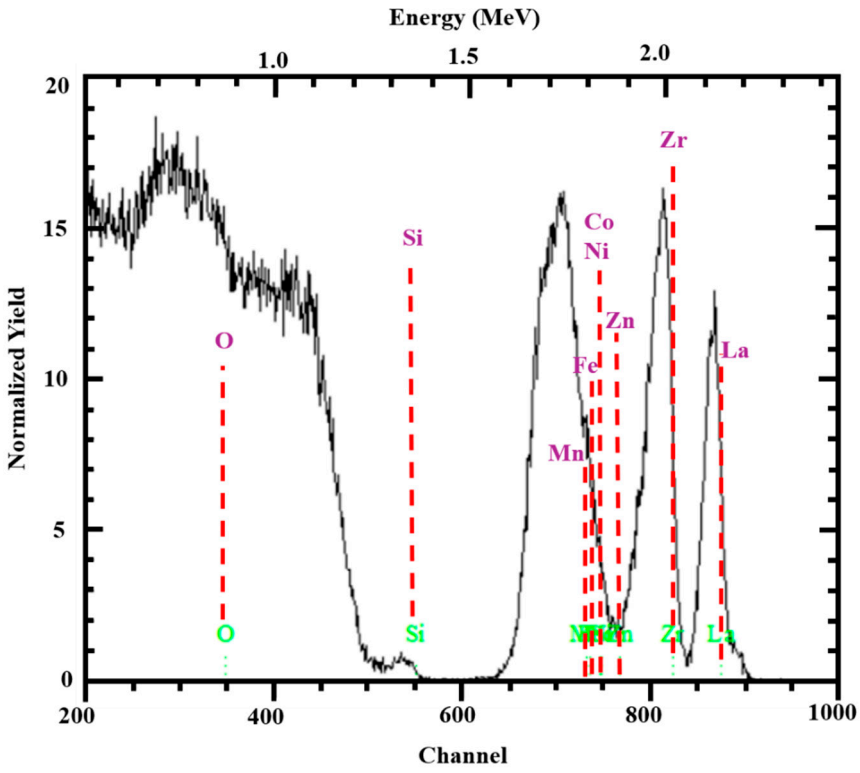


Figure S2: RBS spectra of NLZ100 showing the presence of all elements specific to ZFO, LLZO and NCM layers

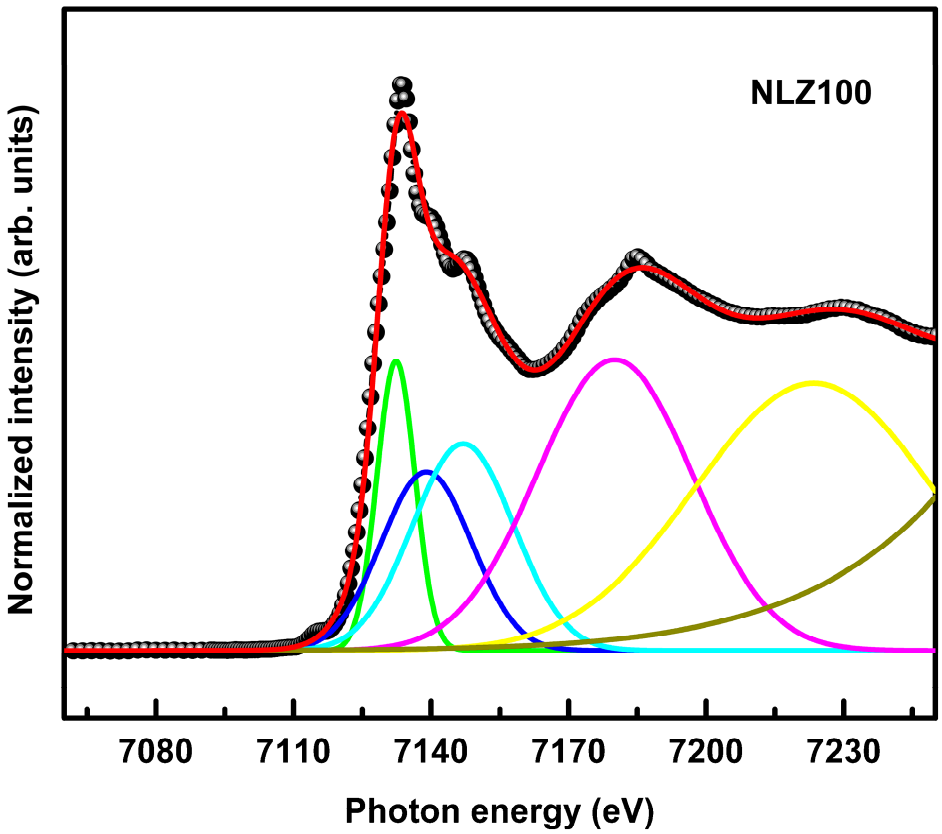


Figure S3: Deconvoluted Fe *K*-edge XANES spectra for the NLZ100 trilayer structure.

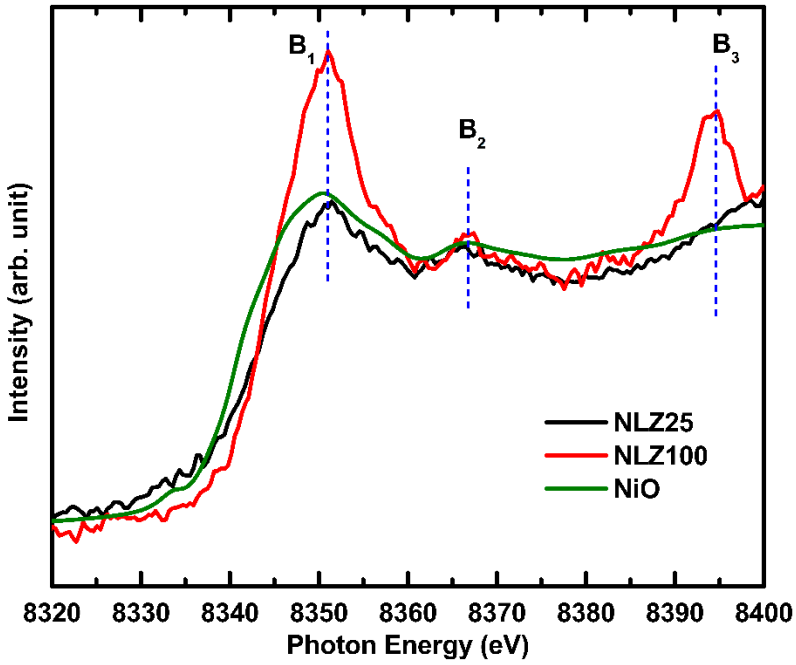


Figure S4: Ni *K*-edge XANES spectra of NLZ25, NLZ100 trilayer structures alongwith NiO reference.

Table S1. Raman bands and their assignments for ZFO.

Assignments	Raman Shift (Cm^{-1})		
	Nanoparticles [39]	Thin Films	
		RF sputtering [40]	Pulsed laser deposition [41]
F_{2g} (1)	221	-	-
E_g	246	-	286
F_{2g} (2)	355	322 - 336	-
F_{2g} (3)	451	474 - 482	487
A_{1g}	647	639 - 664	613

Table S2. Raman bands and their assignments corresponding to LLZO.

Assignments	Raman Shift (Cm^{-1})		
	Cubic Garnet structure		Tetragonal Garnet structure
	Nanoparticles [42]	Single Crystal [43]	Single Crystal [43]
T_{2g}	-	-	100
T_{2g}	107	107	108
E_g	121	121	122
E_g	-	-	127
T_{2g}	-	144	157
E_g	-	-	183
T_{2g}	209	209	206
T_{2g}	-	-	228
A_{1g}	251	251	246
T_{2g}	-	-	288
E_g	-	-	336
	-	-	356
T_{2g}	361	361	362
A_{1g}	-	-	376
E_g or T_{2g}	410	410	402
T_{2g}	-	-	425
E_g	-	-	442
T_{2g}	-	-	484
	-	-	496
T_{2g} or E_g	514	514	517
E_g	-	-	595
A_{1g}	-	645	642
T_{2g}	-	-	712

Table S3. Raman bands and their assignments for NCM.

Assignments	Raman Shift (Cm^{-1})	
	Ni-Rich $Li_xNi_{1-2y}Co_yMn_yO_2$ [44]	**Silver-coated $LiNi_{0.8}Co_{0.1}Mn_{0.1}O_2$ [45]**
	390	-
E_g	463	475
	502	554
A_{1g}	566	586
	609	-

Table S4. ICP-MS analysis of NLZ100.

Sample name	^{7}Li (no gas used)		^{7}Li (He gas used)	
	Conc. (ppb)	**Conc. RSD**	**Conc. (ppt)**	**Conc. RSD**
NLZ100 (diluted)	4.383	3.6	5.063	3.1
NLZ100 (undiluted)	36.341	0.8	43.563	3.7

Weight (m) of the sample = 0.1452 gm
Volume (v) = 15 mL
Dilution factor, f = 1 and 10

Table S5. Composition and thickness of NLZ25 and NLZ100.

NLZ25		
Layer	**Composition**	**Thickness (nm)**
1	$Li_{0.81}Ni_{0.7}Mn_{0.05}Co_{0.01}O_3$	30
2	$La_{0.2}Zr_{2.5}O_1$	10
3	$Li_{2.1}La_{0.5}Zr_5O_3$	57
4	$La_{0.15}Fe_{0.1}O_1$	10
5	$Zn_{0.1}Fe_{0.8}O_3$	70
6	$Ni_{0.05}Fe_{0.15}La_{0.1}Zn_{0.1}Si_1O_2$	130
7	Si_1O_2	300
8	Si_1	10000
NLZ100		
1	$Li_{0.84}Ni_{0.7}Mn_{0.1}Co_{0.05}O_1$	40
2	$La_{0.6}Zr_{0.5}Zn_{0.7}O_1$	10
3	$Li_{3.9}La_{0.5}Zr_{1.5}O_4$	90
4	$La_{0.1}Fe_{0.01}Zr_{0.65}O_2$	20
5	$Zn_{0.85}Fe_{1.9}O_3$	200
6	$Fe_{0.2}Zn_{0.05}Si_1O_2$	90
7	$La_{0.07}Fe_{0.02}Si_1Zn_{0.05}O_2$	150
8	Si_1	10000