Supporting Information

Reuse, Recycle and Regeneration of LiFePO₄ Cathode from Spent Lithium-ion Batteries for Rechargeable Lithium and Sodium-ion Storage

Binitha Gangaja†, Shantikumar Nair† and Dhamodaran Santhanagopalan†*

[†]Centre for Nanosciences, Amrita Vishwa Vidyapeetham, AIMS (P.O), Kochi – 682 041, India.

*Corresponding Author email: dsgopalan20710@aims.amrita.edu

Total number of pages: 12 (S1 – S12)

Number of figures: 17 (S1-S17)

Number of Tables: 1 (Table S1)

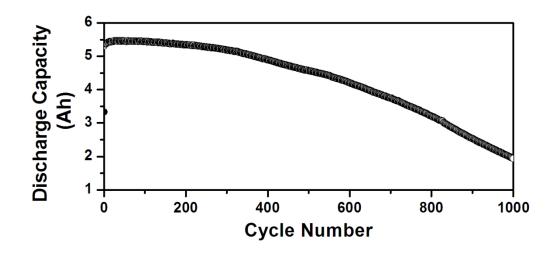


Figure S1. Cycling performance of the commercial cell for 1000 cycles at 5A current rate.

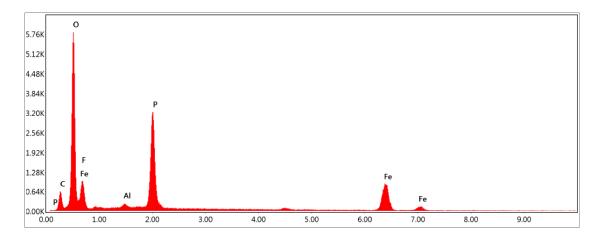


Figure S2. EDX spectra showing the presence of element Fe, P, O, F, C and Al.

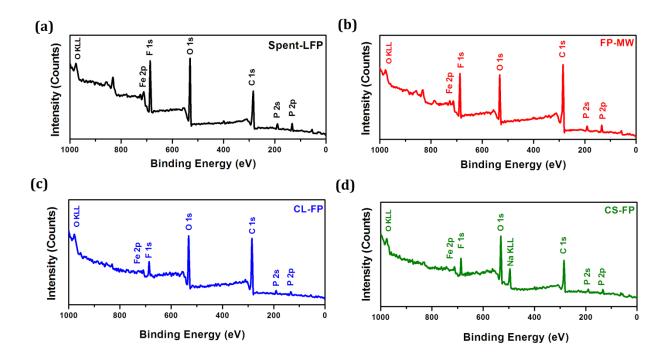


Figure S3. XPS survey spectrum (a) spent LFP, (b) FP-MW, (c) CL-FP and (d) CS-FP.

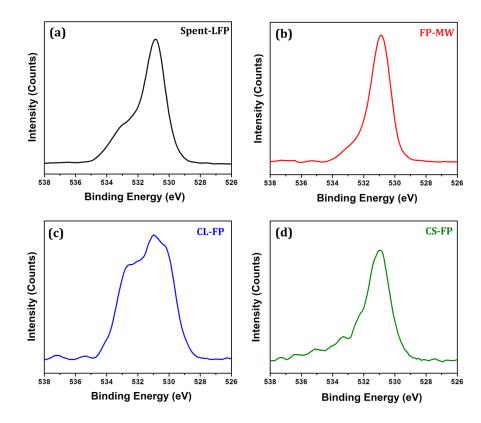


Figure S4. High resolution O 1s spectrum of (a) spent LFP, (b) FP-MW, (c) CL-FP and(d) CS-FP.

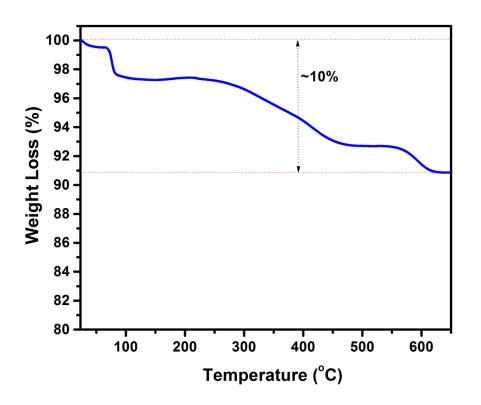


Figure S5. TGA analysis of the spent LFP electrode from 23°C to 650°C in air.

Table S1. Electrochemical performance comparison of fresh LiFePO4 cathodes available in
literature with reused and recycled LFP cells

SI No.	LiFePO ₄ Sample details	C-Rate	Capacity (mAh/g)	Reference*
1	LFP	C/5	110	
		1C	80	1
	LFP + 1% Cu	C/5	140	
		1C	100	
2	C-LFP	C/10	145	
		10C	50	2
	C-LFP/7% Polypyrrole	C/10	148	
		10C	100	
3	LFP Bare	C/10	146	
		5C	90	3
	LFP Surfactant	C/10	170	
		5C	142	

4	LFP	C/10	132	
		5C	83.4	4
5	LFP	C/10	110	
5		C/10	118	
	F-Doped LFP	C/10	175	5
6	LFP	C/10	140	
		10C	70	6
			-	0
	LFP/Nanofibers	C/10	150	
		10C	80	
7	LFP @ Glucose	C/5	160	
		10C	100	7
8	Commercial LFP	1C	130	
-		30C	60	0
		500	00	8
	Self-assembled LFP	1C	150	
		30C	110	
9	Recycled LFP	1C	150	This work
		10C	95	
10	CL-FP	1C	145	This work
		10C	107	

^{*}References included at the end

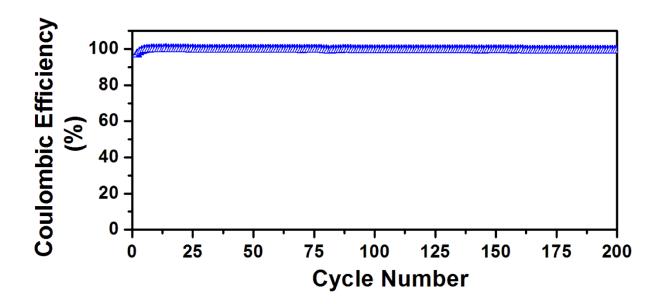


Figure S6. Coulombic Efficiency plot of reused spent-LFP LIB cell at 5C rate.

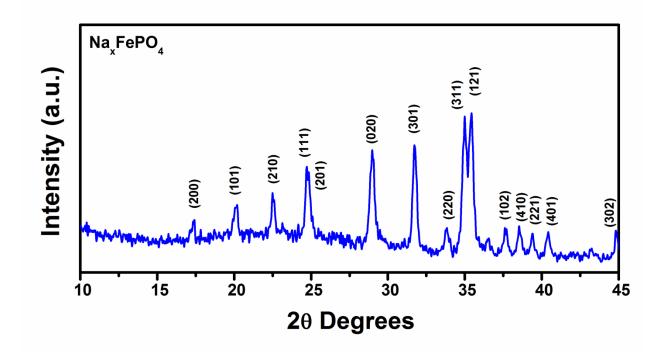


Figure S7. X-Ray diffraction pattern of electrochemically sodiated Na_xFePO₄.

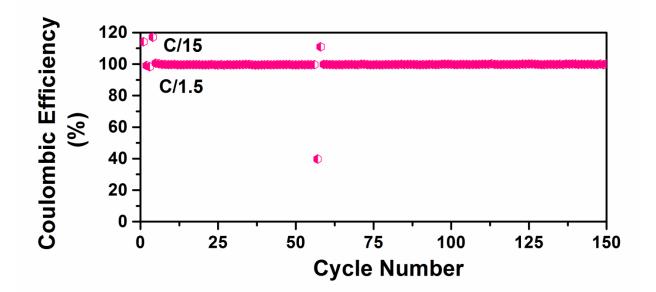


Figure S8. Coulombic Efficiency plot of MW processed FP-NIB cell, first 3 cycles at C/15 and then at C/1.5.

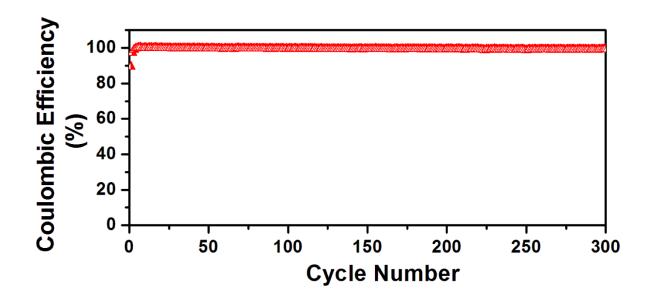


Figure S9. Coulombic Efficiency plot of LiI-FP LIB cell at 5C rate.

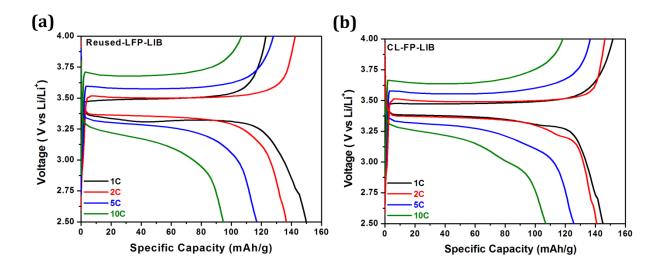


Figure S10: Lithium ion battery charge-discharge profiles of (a) reused-LIB and (b) CL-FP electrodes at different rates in the potential window from 2.5 V to 4V.

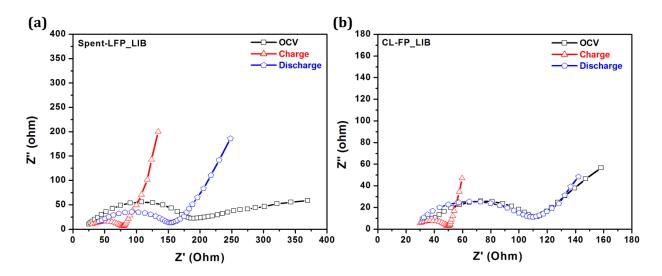


Figure S11: Electrochemical impedance analysis of (a) Spent-LFP and (b) CL-FP lithium ion cells in the frequency range from 10 kHz to 10 mHz at three different stages as indicated in legend.

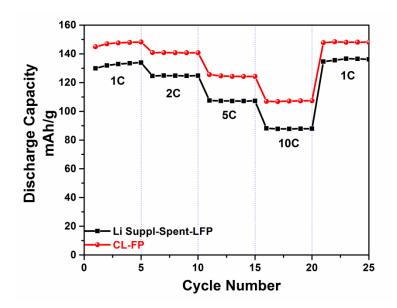


Figure S12: Rate performance comparison plot of lithium supplemented Spent-LFP and chemically lithiated FP.

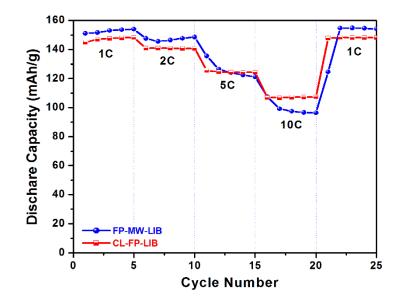


Figure S13: Rate performance plot of microwaved FP electrode (FP-MW) for lithium ion battery in comparison of chemically lithiated FP electrode. Rate performances were done at different rates (as shown in legend) for 5 cycles in the potential window from 2.5 V to 4V.

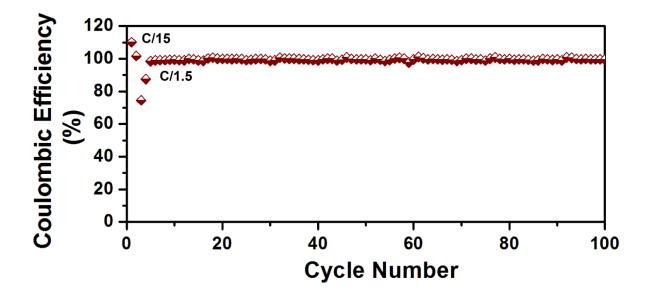


Figure S14. Coulombic Efficiency plot of NaI-FP sodium ion cell, first 3 cycles at C/15 and following cycles at C/1.5.

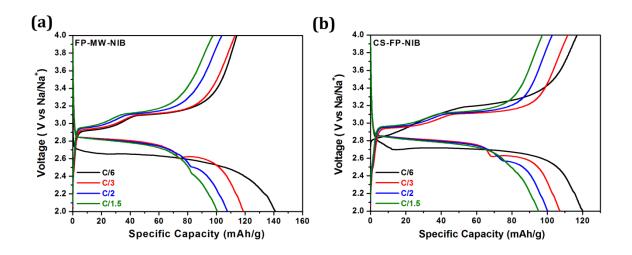


Figure S15: Sodium ion battery charge-discharge profiles of (a) FP-MW and (b) CS-FP electrodes at different rates from C/6 to C/1.5 as mentioned in the legend.

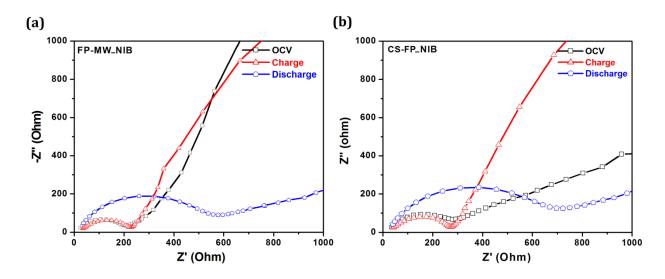


Figure S16: Electrochemical impedance analysis of (a) FP-MW and (b) CS-FP sodium ion cells in the frequency range from 10 kHz to 10 mHz at three different stages as indicated in legend.

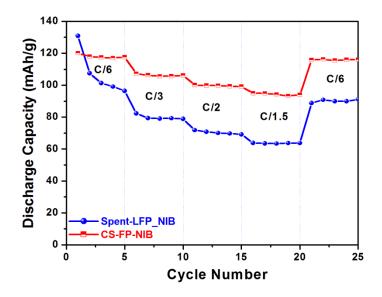


Figure S17: Rate performance plot of Spent-LFP electrode for sodium ion battery applications in comparison with CS-FP-NIB. The electrodes were cycled at different rates (as shown in legend) for 5 cycles in the potential window from 2.0 V to 4.0 V.

References:

- Croce, F.; D'Epifanio, A.; Hassoun, J.; Deptula, A.; Olczac, T.; Scrosati, B. A Novel Concept for the Synthesis of an Improved LiFePO₄ Lithium Battery Cathode. *Electrochem. Solid-State Lett.* 2002, 5 (3), 47–50, DOI 10.1149/1.1449302.
- (2) Huang, Y. H.; Goodenough, J. B. High-Rate LiFePO₄ Lithium Rechargeable Battery Promoted by Electrochemically Active Polymers. *Chem. Mater.* 2008., 20 (23), 7237–7241, DOI 10.1021/cm8012304.
- (3) Choi, D.; Kumta, P. N. Surfactant Based Sol-Gel Approach to Nanostructured LiFePO₄ for High Rate Li-Ion Batteries. *J. Power Sources* 2007, *163* (2), 1064–1069, DOI 10.1016/j.jpowsour.2006.09.082.
- (4) Liu, Y.; Gu, Y.-J.; Luo, G.-Y.; Chen, Z.-L.; Wu, F.-Z.; Dai, X.-Y.; Mai, Y.; Li, J.-Q. Ni-Doped LiFePO₄/C as High-Performance Cathode Composites for Li-Ion Batteries. *Ceram. Int.* 2020, 46 (10), 14857–14863, DOI 10.1016/j.ceramint.2020.03.011.
- (5) Wang, X.; Feng, Z.; Hou, X.; Liu, L.; He, M.; He, X.; Huang, J.; Wen, Z. Fluorine Doped Carbon Coating of LiFePO₄ as a Cathode Material for Lithium-Ion Batteries. *Chem. Eng. J.* **2020**, *379* (July 2019), 122371, DOI 10.1016/j.cej.2019.122371.
- (6) Adepoju, A. A.; Williams, Q. L. High C-Rate Performance of LiFePO₄/Carbon Nanofibers Composite Cathode for Li-Ion Batteries. *Curr. Appl. Phys.* 2020, 20 (1), 1–4, DOI 10.1016/j.cap.2019.09.014.
- (7) Yuan, M.; Li, Y.; Zhang, K.; Li, Y.; Yao, Y. One-Step Liquid Phase Synthesis of LiFePO₄@C

Composite as High Performance Cathode Material for Lithium-Ion Batteries. *Nano* **2020**, *15* (06), 2050080, DOI 10.1142/S1793292020500800.

(8) Peng, L.; Zhao, Y.; Ding, Y.; Yu, G. Self-Assembled LiFePO₄ Nanowires with High Rate Capability for Li-Ion Batteries. *Chem. Commun.* 2014, *50* (67), 9569–9572, DOI 10.1039/c4cc04036h.