

## ELECTRONIC SUPPLEMENTARY INFORMATION

### Preliminary Challenges in Upscaling the Recovery of Lithium from Spent Lithium-ion Batteries by Electrochemical Method: A Review

Mohamad Arif Kasri<sup>1,2</sup>, Muhammad Zharfan Mohd Halizan<sup>2</sup>, Irina Harun<sup>3</sup>, Fadzli Irwan Bahrudin<sup>4</sup>, Nuraini Daud<sup>5</sup>, Muhammad Faiz Aizamddin<sup>6</sup>, Siti Nur Amira Shaffee<sup>6</sup>, Norazah Abd Rahman<sup>7</sup>, Saiful Arifin Shafiee<sup>1\*</sup>, Mohd Muzamir Mahat<sup>2\*</sup>

**Table S1.** Preliminary economic analysis for recovery of one-ton scrap  $\text{LiMn}_2\text{O}_4$  by slurry electrolysis. <sup>1</sup>

<b>Revenue</b>	<b>Products</b>	<b>Mass (kg)</b>	<b>Price (USD/kg)</b>	<b>Benefits (\$)</b>
	$\text{Li}_2\text{CO}_3$	92.74	12.00	1112.88
	$\text{MnO}_2$	441.42	2.20	971.12
	$\text{Na}_2\text{SO}_4 \cdot 10\text{H}_2\text{O}$	1965.00	0.09	176.85
	<b>Total</b>	-		<b>2260.85</b>
<b>Energy cost</b>	<b>Procedure</b>	<b>Energy consumption (kWh)</b>	<b>Price (USD*kWh)</b>	<b>Cost (\$)</b>
	Slurry electrolysis	1173.33	0.15	-176.00
	Filtering, drying and sieving	230	0.15	-34.50
	Evaporative crystallization	1100	0.15	-165.00
	Freeze crystallisation	110	0.15	-16.50
	<b>Total</b>	2613.33	0.15	<b>-392.00</b>
<b>Reagent cost</b>	<b>Reagents</b>	<b>Dosage (kg)</b>	<b>Price (USD/kg)</b>	<b>Cost (\$)</b>
	NaOH	488.20	0.60	-292.92
	$\text{H}_2\text{SO}_4$	598.05	0.25	-149.51
	<b>Total</b>			<b>-442.43</b>
<b>Net profit</b>				<b>1426.42</b>

**Table S2.** Preliminary economic analysis for recovery of one-ton scrap  $\text{LiMn}_2\text{O}_4$  by various methods. <sup>1</sup>

Process	Reagent cost (USD)	Energy cost (USD)	Revenue	Profit	Reference
Pyrometallurgy	-	559.05	1687.14	1128.09	2
Hydrometallurgy	-	387.53	1237.78	850.25	3
Hydrometallurgy	659.28	415.5	1303.10	228.32	4
Electrochemical (slurry electrolysis)	442.43	392.00	2260.85	1426.42	1

**Table S3.** Economic analysis for recycling one ton of spent LiFePO<sub>4</sub> batteries by the electrochemical method (suspension electrodialysis) in China (exchange rate: 1 USD = 6.72 CNY).<sup>5</sup>

<b>Revenue</b>	<b>Products</b>	<b>Mass (kg)</b>	<b>Price (USD/kg)</b>	<b>Benefits (USD)</b>
	LiOH·H <sub>2</sub> O	84.28	15.00 <sup>10</sup>	1264.20
	FePO <sub>4</sub>	302.45	1.00 <sup>11</sup>	302.45
	<b>Total</b>		-	<b>1566.65</b>
<b>Energy cost</b>	<b>Procedure</b>	<b>Energy consumption (kWh)</b>	<b>Price (USD*kWh)</b>	<b>Cost (USD)</b>
	Suspension electrodialysis	1742.40	0.15	-261.36
	Filtering, drying and sieving	230.00	0.15	-34.50
	Evaporative crystallisation	802.67	0.15	-120.40
	<b>Total</b>		-	<b>-416.26</b>
<b>Net profit</b>				<b>1150.39</b>

**Table S4.** Preliminary economic analysis for recovery of one-ton spent LFP batteries in various methods.<sup>5</sup>

<b>Products</b>	<b>Reagent cost (USD)</b>	<b>Energy cost (USD)</b>	<b>Revenue (USD)</b>	<b>Profit (USD)</b>	<b>References</b>
Li <sub>2</sub> CO <sub>3</sub> , FePO <sub>4</sub>	275.70	219.00	1272.2	777.5	6
Li <sub>2</sub> CO <sub>3</sub> , FePO <sub>4</sub>	274.22	106.50	1329.90	949.18	7
Li <sub>3</sub> PO <sub>4</sub> , FeC <sub>2</sub> O <sub>4</sub> ·2H <sub>2</sub> O	858.00	77.88	1901.13	965.25	8
LiOH·H <sub>2</sub> O, FePO <sub>4</sub>	-	416.26	1566.65	1150.39	5

## REFERENCES

1. Li, Z., He, L., Zhao, Z. wei, Wang, D. & Xu, W. Recovery of Lithium and Manganese from Scrap  $\text{LiMn}_2\text{O}_4$  by Slurry Electrolysis. *ACS Sustain Chem Eng* **7**, 16738–16746 (2019).
2. Xiao, J., Li, J. & Xu, Z. Recycling metals from lithium ion battery by mechanical separation and vacuum metallurgy. *J Hazard Mater* **338**, 124–131 (2017).
3. Kondás, J., Jandová, J. & Nemeckova, M. Processing of spent Li/MnO<sub>2</sub> batteries to obtain  $\text{Li}_2\text{CO}_3$ . *Hydrometallurgy* **84**, 247–249 (2006).
4. Meshram, P., Abhilash, Pandey, B. D., Mankhand, T. R. & Deveci, H. Comparison of Different Reductants in Leaching of Spent Lithium Ion Batteries. *JOM* **68**, 2613–2623 (2016).
5. Li, Z., He, L., Zhu, Y. & Yang, C. A Green and Cost-Effective Method for Production of LiOH from Spent  $\text{LiFePO}_4$ . *ACS Sustain Chem Eng* **8**, 15915–15926 (2020).
6. Zhang, J. *et al.* Sustainable and Facile Method for the Selective Recovery of Lithium from Cathode Scrap of Spent  $\text{LiFePO}_4$  Batteries. *ACS Sustain Chem Eng* **7**, 5626–5631 (2019).
7. Yang, Y. *et al.* A Closed-Loop Process for Selective Metal Recovery from Spent Lithium Iron Phosphate Batteries through Mechanochemical Activation. *ACS Sustain Chem Eng* **5**, 9972–9980 (2017).
8. Fan, E. *et al.* Selective Recovery of Li and Fe from Spent Lithium-Ion Batteries by an Environmentally Friendly Mechanochemical Approach. *ACS Sustain Chem Eng* **6**, 11029–11035 (2018).