Electronic Supplementary Information

Separation and recovery of graphite from spent lithium-ion batteries

for synthesizing micro-expanded sorbents

Zhiwei Geng^{a,b,1}, Junjie Liu^{a,b,1}, Yanni Geng^{a,b}, Mingming Peng^{a,b}, Mopeng Xiong^{a,b},

Hui Shi^{a,b,*} and Xubiao Luo^{a,b}

This paper is preparing for publication in New Journal of Chemistry

Section 1: Economic and Environmental Benefit Assessment

Based on the data provided by reference, and introducing the EverBatt model¹ and the GREET model,² we performed a laboratory-scale economic and environmental analysis^{3, 4} of spent graphite for the preparation of adsorbents. Assume that 1 kg of NCM111 battery waste is recycled. The battery cost is 0.00\$/kg based on donation, and the weight of graphite in the battery is 19.4%,⁵ corresponding to 194g. Do not consider the pretreatment cost of spent graphite recovery such as battery dismantling, as well as equipment cost and other costs, only focus on the process of preparing adsorbent from spent graphite. In addition, the calculation of reagent cost and equipment energy consumption is based on the manufacturer in this article. The electric furnace power is 2.5kW, the constant temperature magnetic stirring power is 840W, and the electricity fee is 0.23\$/kWh.⁶ Taking into account the product loss during the preparation of the adsorbent, the recovery rate is calculated at 90%, and the price of the product is based on the price of similar products. Furthermore, does not take into account the loss of CO₂, the process CO₂ emissions from battery recycling are calculated as^{1.7}

$$P_{CO2} = P_{CO2,combusion} + P_{CO2,decomposition} \tag{1}$$

Where $P_{CO2,combustion}$ denotes process CO_2 emissions from material combustion, $P_{CO2,decomposition}$ represents process emissions from material decomposition during recycling processes. $P_{CO2,decomposition}$ is estimated from stoichiometry, and $P_{CO2,combustion}$ can be estimated as

$$P_{CO2,combusion} = \sum_{i} m_i \times \frac{Carbon \ content_i}{Carbon \ content_{CO2}}$$
(2)

Where m_i denotes the mass of material i that is combusted in the recycling process, and Carbon content_i denotes the carbon content of material i, estimated in molar mass.

The economic and environmental benefit analysis are shown in Table S1, calculated at the exchange rate of RMB to US dollar of 1\$=7.0129 RMB.



Fig. S1 (a) Nitrogen adsorption-desorption isotherms, (b) pore size distribution,

and (c) contribution of pore size to pore volume of SG and MEG.

Section 3:

Samples	Specific surface area (m ² /g)	Pore volume (cm^3/g)	Pore size (nm)
SG	2.2779	0.003581	41.4662
MEG	2.6501	0.007414	28.4889

Table S1. Pore system of SG and MEG.

Types	ID	I _G ,	$I_D \ / \ I_{G'}$
SG	637455	414799	1.537
MEG	566896	409855	1.383

Table S2. Pore system of SG and MEG.

Adsorbents	Cost (\$/kg cell)	Energy consumption (MJ/kg)	Economic effects (\$/kg cell)	Greenhouse gases emission (g/kg)	Ref.
MnO ₂ -AG	46.48	66.312	-31.54	640.44	Tuo Zhao et al., 2017 ⁸
RTEG	74.75	_	-44.87	639.56	Ting Liu et al., 2017 ⁹
Mg-MCMB	57.54	29.646	-45.47	651.88	Yan Zhang et al., 2016 ¹⁰
MEG	26.68	26.388	3.2	639.56	This study

Table S3. Economic and environmental benefit analysis for recycling spent

graphite to prepare sorbents

References

1. S. J. Dai Q, Ahmed S, et al., *Argonne National Lab.(ANL), Argonne, IL (United States)*, 2019, DOI: https://doi.org/10.2172/1530874.

2. E. A. Wang M, Lee U, et al., Argonne National Lab.(ANL), Argonne, IL (United States), 2021.

3. M. Li, B. Zhang, X. Qu, M. Cai, D. Liu, F. Zhou, H. Xie, S. Gao and H. Yin, *ACS Sustainable Chemistry & Engineering*, 2022, **10**, 8305-8313.

4. J. Wang, Q. Zhang, J. Sheng, Z. Liang, J. Ma, Y. Chen, G. Zhou and H. M. Cheng, *Natl Sci Rev*, 2022, **9**, nwac097.

5. G. L. Dunn J B, Barnes M, et al., *Argonne National Lab.(ANL), Argonne, IL (United States)*, 2014, DOI: <u>https://doi.org/10.2172/1177517</u>.

6. Y. Huang, P. Shao, L. Yang, Y. Zheng, Z. Sun, L. Fang, W. Lv, Z. Yao, L. Wang and X. Luo, *Resources, Conservation and Recycling*, 2021, **174**.

7. M. Mohr, J. F. Peters, M. Baumann and M. Weil, *Journal of Industrial Ecology*, 2020, **24**, 1310-1322.

8. T. Zhao, Y. Yao, M. Wang, R. Chen, Y. Yu, F. Wu and C. Zhang, *ACS Appl Mater Interfaces*, 2017, **9**, 25369-25376.

T. Liu, R. Zhang, X. Zhang, K. Liu, Y. Liu and P. Yan, *Carbon*, 2017, **119**, 544-547.
Y. Zhang, X. Guo, F. Wu, Y. Yao, Y. Yuan, X. Bi, X. Luo, R. Shahbazian-Yassar, C. Zhang and K. Amine, *ACS Appl Mater Interfaces*, 2016, **8**, 21315-21325.