



## Editorial

## Direct recycling of Lithium ion battery, what's next?

## ARTICLE INFO

## Keywords:

Li-ion battery recycling

The energy transition posed new technological challenges due to the increasing need for energy storage systems in electric mobility and renewable energies. The current reference technology for energy storage in electronics, electric vehicles, and stationary accumulation is represented by Li-ion batteries, whose market is expected to grow rapidly with interesting prospects for new production players located in Western countries [1].

However, these prospects of expansion may be hampered by some critical issues in the supply of raw materials necessary for the manufacturing of Li-ion batteries and the end-of-life treatment of these devices. In fact, even if the secondary raw materials recovered by battery recycling will only be able to provide a partial contribution to the supply of battery raw materials [2], the recycling of end-of-life batteries is mandatory to guarantee the environmental sustainability of the energy transition by avoiding both the loss of resources and environmental pollution.

The processes for recycling Li-ion batteries at the end of their life can be different [3], each characterized by limitations that currently hinder the application at large scale. End-of-life Li-ion batteries are characterized by a complex structure and are heterogeneous from a chemical point of view due to the continuous evolution of the cathode materials.

The currently most applied recycling approach includes the recourse to pyrometallurgical processes, which do not require pre-treatment of batteries, but are energy intensive and allow for the recovery of only the most noble metals (Cu, Co, Ni) in alloy form, thus not providing a direct material supply to the battery-manufacturing sector. The alternative is represented by hydrometallurgical processes, which require the development of pre-treatment processes for the separation of the electrode material, subsequently subjected to metal dissolution, purification operations, and synthesis of precursors suitable for the production of cathode materials. These latter processes require lower energy consumption compared to pyrometallurgical ones and allow, in principle, for the recovery of all the battery components (including Li, graphite and non-metals), but they can be extremely complex and costly, thus not economically sustainable, considering the current collection volumes of end-of-life Li-ion batteries.

Direct recycling [4] is the third new alternative. In this case, after the separation of the different battery components by physical pre-treatment, chemical or thermal regeneration of the recovered cathode powder is performed. In this way, the direct re-constitution of the cathode materials is attained, avoiding the recourse to the complex and costly cathode

dissolution, and salt or precursor recovery by hydrometallurgical operations. From this perspective, direct recycling configures a competitive technological alternative for the treatment of end-of-life Li-ion batteries [5], holding the potential to achieve a significantly lower processing cost, improved environmental impact, and a better integration into a circular business model as compared to state-of-the-art pyrometallurgical and hydrometallurgical recycling processes.

On the other hand, new technological challenges are introduced by the large-scale implementation of direct recycling in relation to battery sorting, liberation, separation of components, and regeneration of cathode materials.

To regenerate cathode materials, end-of-life Li-ion batteries have to be accurately sorted according to the cathode chemistry, and then the cathode materials should be liberated from the other battery components with a degree of purity high enough not to affect the electrochemical performances of the materials after regeneration. Finally, methodologies allowing to restore efficiently the structure and composition of the cathode materials have to be developed.

Some of these treatments are in a very early stage, as the sorting of waste flux with respect to the chemistry of Li-ion batteries. The development of “chemistry sensitive” sorting could be facilitated by the implementation of policies aimed at favouring the development of recycling processes, such as the labelling system envisaged by the Battery Proposal 2020 for batteries placed on the EU market starting from 2030 [6].

Other treatments, such as dismantling by comminution, are already implemented in industrial practice for the recovery of electrode materials (black mass) fed to hydro- and pyrometallurgical processes. However, even these treatments require further optimization to enable the development of direct recycling processes, specifically considering the effects that fine grinding can have not only in terms of component liberation, but also in terms of impurities present in the subsequently recovered electrode material [7].

Likewise, the physical pre-treatment methods currently available for the recovery of cathode and anode material fractions require further optimization and upgrading as well as their integration with innovative pre-treatment stages in order to reach the requirements of separation efficiency and purity required by direct recycling. Physical separation methods and enrichment treatments of the cathode material based on conventional ore dressing techniques, such as flotation, can pave the

way towards direct recycling along with more innovative technologies (such as optical sorting) that are currently used for other technological wastes, but may be extended and adapted for Li-ion batteries. In this framework, the development of innovative methods for the effective detachment from current collector and the de-agglomeration of the cathode and anode materials can help to decrease the impurities content of the separated electrode materials.

Methods for the efficient regeneration of the recovered cathode materials are then required. Different methods have already been proposed and tested over the past few years confirming the possibility to effectively regenerate the cathode materials from end-of-life Li-ion batteries [8]. However, the performed studies typically included the application of high purity cathode materials manually extracted from the end-of-life batteries. This separation protocol for the recovery of the electrode materials can be hardly applied at industrial scale. Accordingly, working with the cathode materials delivered by physical pre-treatment or purposefully introducing impurities in the treated materials is fundamental to evaluate the feasibility of the proposed regeneration methodologies, allowing, at the same time, to identify the performance requirements that should be targeted by the upstream separation.

Direct recycling could therefore be the sustainable way to recycle lithium ion batteries, but some technological challenges need to be solved for its large-scale implementation. In any case, these technological challenges also constitute important steps for the development of other recovery strategies (such as hydrometallurgical ones) because the better separation of materials, which is a must for direct recycling, could help also the economic sustainability of any other type of process (hydro or pyro) reducing product refining operations and costs.

#### Author contributions

All authors have made contributions to this editorial and approved for its publication.

#### Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

#### References

- [1] A Vision for a Sustainable Battery Value Chain in 2030 Unlocking the Full Potential to Power Sustainable Development and Climate Change Mitigation Insight Report September 2019.
- [2] C. Xu, Q. Dai, L. Gaines, et al., Future material demand for automotive lithium-based batteries, *Commun. Mater.* 1 (2020) 99, <https://doi.org/10.1038/s43246-020-00095-x>
- [3] Wu Xiaoxue, Ma Jun, Wang Junxiong, Zhang Xuan, Zhou Guangmin, Liang Progress Zheng, key issues, and future prospects for Li-Ion battery recycling, *Glob. Chall.* 6 (2022) 2200067, <https://doi.org/10.1002/gch2.202200067>
- [4] Wu Jiawei, Zheng Mengting, Liu Tiefeng, Wang Yao, Liu Yujing, Nai Jianwei, Zhang Liang, Zhang Shanqing, Tao Xinyong, Direct recovery: a sustainable recycling technology for spent lithium-ion battery, *Energy Storage Mater.* Volume 54 (2023) 120–134, <https://doi.org/10.1016/j.ensm.2022.09.029>
- [5] Gaines Linda, Wang Yan, How to maximize the value recovered from Li-ion batteries: hydrometallurgical or direct recycling? *Electrochem. Soc. Interface* 30 (2021) 51, <https://doi.org/10.1149/2.F07213F>
- [6] <<https://eur-lex.europa.eu/legal-content/EN/TXT/HTML/?uri=CELEX:52020PC0798&from=EN>>.
- [7] Kae E. Fink, Bryant J. Polzin, John T. Vaghey, Joshua J. Major, Alison R. Dunlop, Stephen E. Trask, Gerald T. Jeka, Jeffrey S. Spangenberg, Matthew A. Keyser, Influence of metallic contaminants on the electrochemical and thermal behavior of Li-ion electrodes, *J. Power Sources* 518 (2022), <https://doi.org/10.1016/j.jpowsour.2021.230760>
- [8] Gao Hongpeng, Tran Duc, Chen Zheng, Seeking direct cathode regeneration for more efficient lithium-ion battery recycling, *Curr. Opin. Electrochem.* 31 (2022) 100875, <https://doi.org/10.1016/j.coelec.2021.100875>

Francesca Pagnanelli, Pier Giorgio Schiavi, Pietro Altimari  
*Department of Chemistry, La Sapienza University of Rome, P.le Aldo Moro 5, 00185 Rome, Italy*  
*E-mail address:* [francesca.pagnanelli@uniroma1.it](mailto:francesca.pagnanelli@uniroma1.it) (F. Pagnanelli).