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Editorial

New Science Based Concepts for Increased Efficiency in Battery Recycling

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It is a common understanding worldwide that electromobility will have a significant share in passenger transport and that there will be a very dynamic increase in the return volumes of discarded batteries in the future. Whilst, currently, recycling is in the hands of a few and mostly small companies, large companies are increasingly preparing for the circular economy scenario of electric vehicles. This requires robust, safe and efficient processes, on which many research centers are currently working in the international environment. At present, there is no preferred concept for the processing of battery scrap, not to mention any standardization or norming. Politically, the EU specification of a 50% weight-based recovery rate (recycling efficiency, RE) based on cell level is causing widespread discussion. On one hand, such a requirement seems low, but taking into account that 15% and 20% weight shares of electrolytes and graphite, respectively, have not been recoverable to date, this RE alone already poses challenges for many companies. The overall question therefore is whether the holistic weight-based RE is the right way to go, or if element-based quotas for Ni, Co, Cu, Li, etc. are clearly the more target-oriented way [1]. This request has been partly implemented recently by the EU in the new draft of the Battery Recycling Directive [2], where, exemplarily, Li is addressed, with a target of 70% by 2030.

Regardless of this discussion, the recovering of technology elements from Li-based batteries requires mechanical and metallurgical processes in combination. Many options for treating discarded batteries are currently being discussed and investigated. Three exemplary recycling process pathways, A, B, C, are shown as a modular scheme, as the following figure (Figure 1) simplifies. These three process options are already realized at an industrial or at least at a pilot scale and comprise different approaches regarding elements recovered and modules selected.

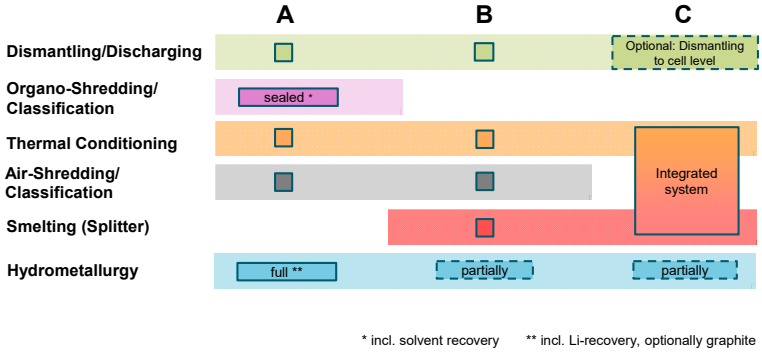

Figure 1. Options and flexibility of battery recycling routes indicating three already industrially applied process paths: **A** (inert shredding and separation before dedicated chemical processing), **B** (thermal conditioning and mechanical separation prior to large-scale production), **C** (direct smelting without pre-conditioning), based on [1].

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It can be seen that different process modules are in place, which can be incorporated into a recycling process. The resulting process A combines inert shredding and mechanical comminution and separation. Thermal conditioning is performed afterwards, whereas process B starts with a thermal treatment and a mechanical comminution and classification is then performed in atmosphere before entering large-scale units. Process C can be inserted into a pyrometallurgical unit with or without dismantling and discharging. Hence, the selection of such modules starting from dismantling/discharging up to hydrometallurgy forms recycling routes A, B, C. Each process path entails specific benefits and drawbacks, for example, based on energy input, eco-footprint or recoverable elements and components. In this scheme, the focus is not set on the economic viability of the main points, but on providing options or scenarios with the focus on the respective technological strengths and weaknesses. Depending on the path taken, the components of a cell can be converted into commodity products, and even raw materials for a closed circle to batteries, or environmentally sensitive substances; Figure 2 is helpful to illustrate the options for obtainable recycling products.

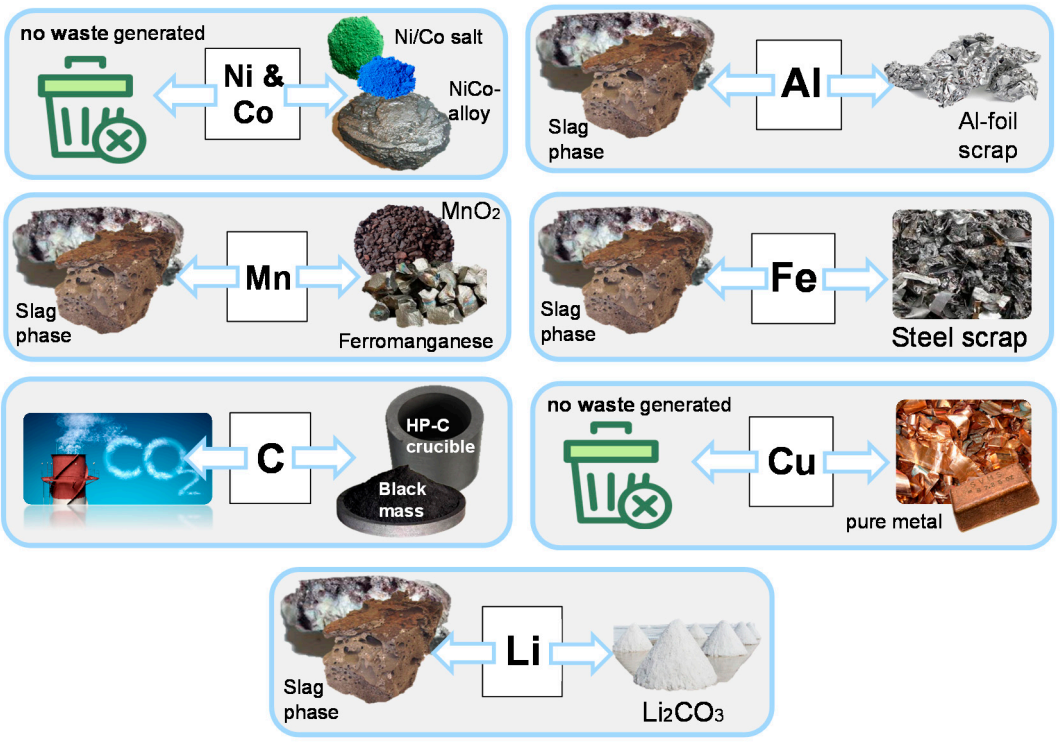

Figure 2. Alternatives of obtained recycling chain products depending on the process paths selected, based on [3]. Here, HP-C refers to high-purity graphite materials used for smelting crucibles.

Noble metals, such as copper (Cu), nickel (Ni) and cobalt (Co), are recovered generally as a marketable and profitable product, independent from the process modules selected. These metals comprise the highest value within a battery, which is why their recovery is a crucial goal in all recycling paths. However, losses in by-products can hardly be avoided since a yield of 100% is practically unrealistic, nevertheless, research always focuses on minimizing process-related losses. Moreover, iron (Fe), represented as steel casings or LFP-cathode systems, and manganese (Mn), represented as an alloying element in steel casings or NMC-cathode systems, are comparatively ignoble metals. Aiming for near zero-waste recycling, Fe and Mn are to be considered as potential products, as well. The mineral phase resulting as slag from pyrometallurgical operations can be transferred to the construction sector, but an elemental recovery of Mn and Fe is not realized in most scenarios. Aluminum (Al) from casing and foils can, similarly to Fe and Mn, be transferred to a slag in a direct

smelting route or recovered to a high degree by advanced mechanical processes, like dismantling, shredding and classification. The graphite in the battery's anodes is a critical element according to the European Union [4], hence, its recovery must play an important role in terms of resource efficiency [5]. Using it as a reducing agent in pyrometallurgical operations is technically possible [6,7] but questionable, since a carbothermic smelting process is under critical view, when aiming at a carbon-free industry approach. Hence, dedicated recovery steps are required. Lithium (Li) represents the mainstay of proven and all innovative battery systems, and its recovery always requires hydrometallurgical operations. Lithium carbonate, but also lithium hydroxide, are marketable products, whose production in recycling ensures the principle of a circular economy.

The obtainable products correspond to a wide range of processes alternatives, which has already been outlined by the different process combinations A, B, C. The established large-scale operations for comparable commodity materials (outer ring) are certainly the most economic ones and, thus, it should be a battery recycling process target to reach an entry point into these production levels as fast as possible. As a matter of fact, the available options are highly diversified, leading to more recycling process alternatives. Figure 3 visualizes this by showing most of the battery recycling options as a modular wheel.

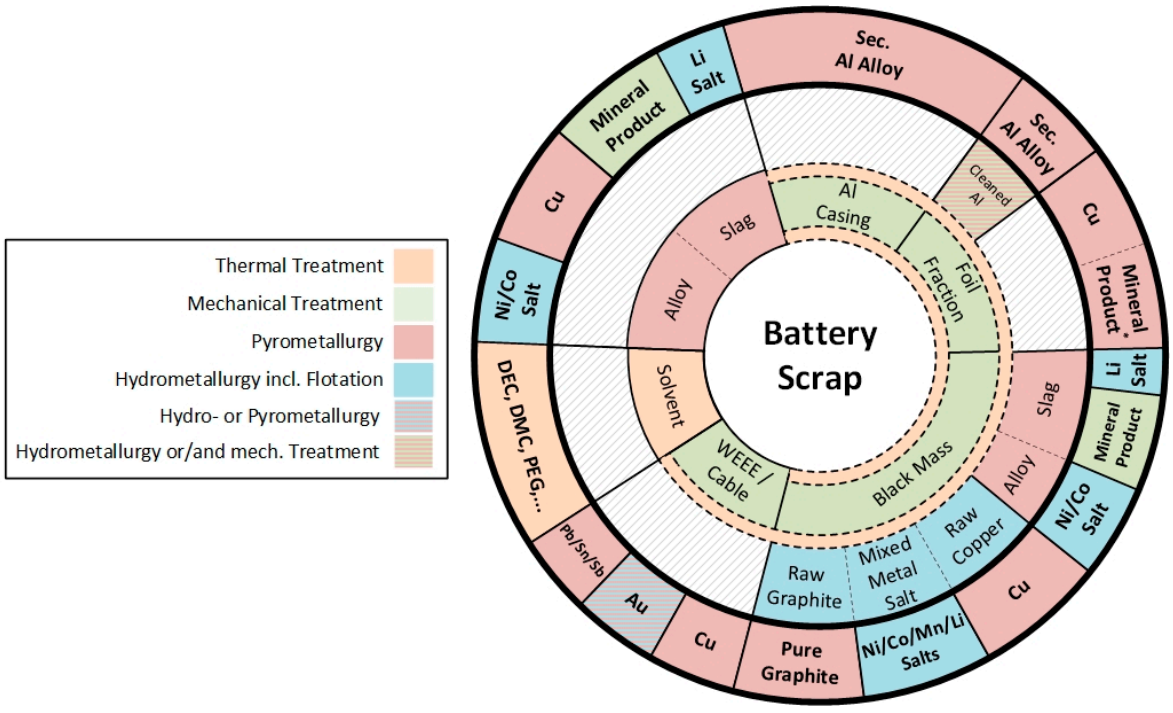

Figure 3. Entry points for recycling products in current large-scale metal production value chains, based on different process modules, based on [8]. Here, the outer ring represents established large-scale operations for comparable commodity materials, also indicated as base material production. The middle ring stands for an advanced second stage in dedicated battery recycling, whereas the inner ring stands for the first process stage in dedicated battery recycling.

This wheel can be read starting from the center with end-of-life batteries, passing two rings of first and possibly second dedicated battery recycling steps into the commodity material production systems (outer ring). Hence, the two inner rings refer to battery-specific recycling facilities and unit operations, and the outer ring represents existing, large-scale metallurgical or chemical industrial production facilities. Obtained products from dedicated battery recycling facilities are channeled into existing large-scale operations for new product generation. Battery scrap, which can be either cell or module based, is treated in process modules, which obtain a specific product fraction. For example, Al casing is recoverable by the module "mechanical treatment", possibly in combination with the module "thermal pre-treatment" before or even after the mechanical process. This Al casing product

is then directly transferred to existing, non-battery-specific aluminum production facilities, where it is transformed to a secondary aluminum alloy by the module "pyrometallurgy".

It is to be pointed out that the entry points of the outer ring are defined in terms of impurities by commodity material producers. Thereby, meeting these demands on battery recycling products is a pre-requisite for generating marketable products. The channeling of battery recycling products into existing infrastructure for primary production is an important tool for near zero-waste recycling. However, with all process chain optimization, it must always be kept in mind that metal losses can also increase with each process module added and that the revenues from the newly recovered products may be offset by the costs of these required additional steps. Additionally, last not but least, all recycling operations must be robust against changing battery chemistries [9,10] and impurities deriving from sorting failures or insufficient orderliness and cleanliness in the plant. One simple example is the future Si-based battery which shows a strong tendency to disable hydrometallurgical operations by gelation risk.

Based on 19 high-quality articles, this Special Issue presents methods for further improving the currently achievable recycling rate, product quality in terms of focused elements and moreover, approaches for the enhanced mobilization of lithium, graphite and electrolyte components. In particular, the target of early stage Li removal is a central point of various research approaches in the world, which has been reported, for example, under the names early stage lithium recovery (ESLR) [11] or CO_2 leaching (COOL) [12]. These processes are a strongly focusing on environmentally friendly lithium mobilization before entering pyrometallurgy or conventional hydrometallurgy. Figure 4 simplifies the effect of this approach.

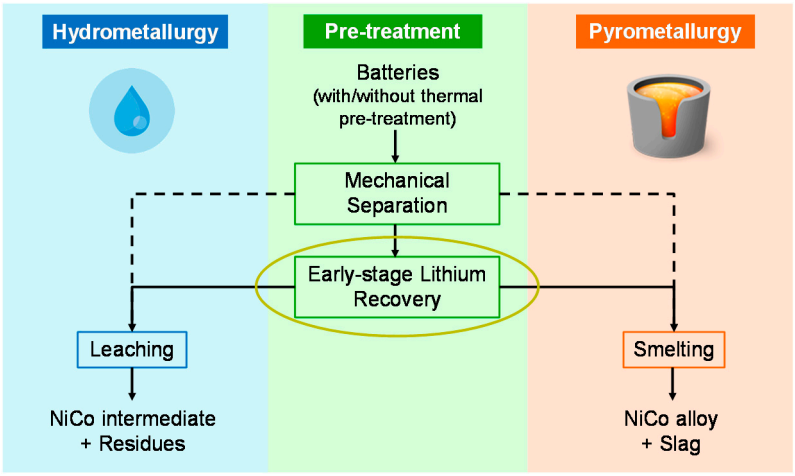

Figure 4. Early stage Li recovery (ESLR) process scheme, based on [11,13].

It has to be pointed out that early stage Li recovery is a tool which can be incorporated into all existing recycling paths, hence, it is an effective add-on for pursuing a high recycling efficiency for this critical element. Currently, the process is investigated by using (thermally treated) black mass [11], but directly shredded material may also be a practical option for future research.

Besides the topic of environmentally friendly lithium mobilization, many more approaches are present in this Special Issue, starting with robotic disassembly and dismantling of Li-ion batteries [14,15]. Moreover, the optimization of various pyro- and hydrometallurgical as well as combined battery recycling processes for the treatment of conventional Li-ion batteries, up to an evaluation of the recycling on an industrial level, and different battery recycling topics, are addressed as well. The recovery of lithium by innovative methods comes to the fore as an important component. In addition to the consideration of the Li distribution in compounds of a Li_2O-MgO-Al_2O_3-SiO_2-CaO system, the Li recovery from battery slags is also discussed. The development of suitable recycling strategies for

new battery systems, such as all-solid-state batteries, but also lithium–sulfur batteries, are also taken into account in this Special Issue. Some articles also discuss the issue that battery recycling processes do not have to produce end products such as high-purity battery materials, but that they should be aimed at finding an "entry point" into existing proven large-scale industrial processes where marketable product generation is possible and cost-efficient (referring to the discussion around Figure 3).

The contributions of this issue are structured according to their research areas, as can be seen in Table 1.

Table 1. Published articles in this Special Issue, "New Science Based Concepts for Increased Efficiency in Battery Recycling" sorted by research field and given as sources in the References.

Research Field	*Source in Special Issue*
Dismantling	[14,15]
Shredding/Separation	[5,16]
Thermal Conditioning	[11]
Smelting	[6,7,17–19]
Hydrometallurgy/Chem. Processing	[9,10,12,20–22]
Reviews	[23–25]

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References

1. Friedrich, B.; Schwich, L. Impact of process chain design on metal recycling efficiency—A critical view on the actual total weight-based target. In Proceedings of the 24th International Congress for Battery Recycling ICBR, Lyon, France, 18–20 September 2019. Available online: https://www.researchgate.net/publication/335889260_Impact_of_process_chain_design_on_metal_recycling_efficiency_-A_critical_view_on_the_actual_total_weight-based_target (accessed on 16 March 2021). [CrossRef]
2. European Commission. Batteries—Modernising EU Rules. Proposal for a Regulation. 2020. Available online: https://ec.europa.eu/info/law/better-regulation/have-your-say/initiatives/12399-Modernising-the-EU-s-batteries-legislation (accessed on 17 March 2021).
3. Friedrich, B.; Schwich, L. Status and Trends of industrialized Li-Ion battery recycling processes with qualitative comparison of economic and environmental impacts. In Proceedings of the 22nd International Congress for Battery Recycling ICBR, Lisbon, Portugal, 20–22 September 2017. Available online: https://www.researchgate.net/publication/319964237_Status_and_Trends_of_industrialized_Li-Ion_battery_recycling_processes_with_qualitative_comparison_of_economic_and_environmental_impacts (accessed on 16 March 2021). [CrossRef]
4. European Commission. Communication from the Commission to the European Parliament, the Council, the European Economic and Social Committee and the Committee of the Regions. On the 2017 List of Critical Raw Materials for the EU. 2017. Available online: https://ec.europa.eu/transparency/regdoc/rep/1/2017/EN/COM-2017-490-F1-EN-MAIN-PART-1.PDF (accessed on 17 March 2021).
5. Ruismäki, R.; Rinne, T.; Dańczak, A.; Taskinen, P.; Serna-Guerrero, R.; Jokilaakso, A. Integrating Flotation and Pyrometallurgy for Recovering Graphite and Valuable Metals from Battery Scrap. *Metals* **2020**, *10*, 680. [CrossRef]
6. Sommerfeld, M.; Vonderstein, C.; Dertmann, C.; Klimko, J.; Oráč, D.; Miškufová, A.; Havlík, T.; Friedrich, B. A Combined Pyro- and Hydrometallurgical Approach to Recycle Pyrolyzed Lithium-Ion Battery Black Mass Part 1: Production of Lithium Concentrates in an Electric Arc Furnace. *Metals* **2020**, *10*, 1069. [CrossRef]
7. Holzer, A.; Windisch-Kern, S.; Ponak, C.; Raupenstrauch, H. A Novel Pyrometallurgical Recycling Process for Lithium-Ion Batteries and Its Application to the Recycling of LCO and LFP. *Metals* **2021**, *11*, 149. [CrossRef]
8. Friedrich, B.; Sabarny, P.; Stallmeister, C. Process Flow Alternatives for LIB Recycling. In *Berliner Recycling- und Sekundärrohstoffkonferenz*; Berlin, Germany, 2021. Available online: https://www.researchgate.net/publication/350071276_Process_Flow_Alternatives_for_LIB_Recycling (accessed on 24 March 2021). [CrossRef]
9. Schwich, L.; Küpers, M.; Finsterbusch, M.; Schreiber, A.; Fattakhova-Rohlfing, D.; Guillon, O.; Friedrich, B. Recycling Strategies for Ceramic All-Solid-State Batteries—Part I: Study on Possible Treatments in Contrast to Li-Ion Battery Recycling. *Metals* **2020**, *10*, 1523. [CrossRef]

10. Schwich, L.; Sabarny, P.; Friedrich, B. Recycling Potential of Lithium–Sulfur Batteries—A First Concept Using Thermal and Hydrometallurgical Methods. *Metals* **2020**, *10*, 1513. [CrossRef]
11. Schwich, L.; Schubert, T.; Friedrich, B. Early-Stage Recovery of Lithium from Tailored Thermal Conditioned Black Mass Part I: Mobilizing Lithium via Supercritical CO2-Carbonation. *Metals* **2021**, *11*, 177. [CrossRef]
12. Pavón, S.; Kaiser, D.; Mende, R.; Bertau, M. The COOL-Process—A Selective Approach for Recycling Lithium Batteries. *Metals* **2021**, *11*, 259. [CrossRef]
13. Stallmeister, C.; Schwich, L.; Friedrich, B. Early-Stage Li-Removal—Vermeidung von Lithiumverlusten im Zuge der Thermischen und Chemischen Recyclingrouten von Batterien. In *Recycling und Rohstoffe*; Holm, O., Thomé-Kozmiensky, E., Goldmann, D., Friedrich, B., Eds.; Thomé-Kozmiensky Verlag GmbH: Neuruppin, Germany, 2020; pp. 545–557. ISBN 9783944310510.
14. Marshall, J.; Gastol, D.; Sommerville, R.; Middleton, B.; Goodship, V.; Kendrick, E. Disassembly of Li Ion Cells—Characterization and Safety Considerations of a Recycling Scheme. *Metals* **2020**, *10*, 773. [CrossRef]
15. Choux, M.; Marti Bigorra, E.; Tyapin, I. Task Planner for Robotic Disassembly of Electric Vehicle Battery Pack. *Metals* **2021**, *11*, 387. [CrossRef]
16. Sinn, T.; Flegler, A.; Wolf, A.; Stübinger, T.; Witt, W.; Nirschl, H.; Gleiß, M. Investigation of Centrifugal Fractionation with Time-Dependent Process Parameters as a New Approach Contributing to the Direct Recycling of Lithium-Ion Battery Components. *Metals* **2020**, *10*, 1617. [CrossRef]
17. Schirmer, T.; Qiu, H.; Li, H.; Goldmann, D.; Fischlschweiger, M. Li-Distribution in Compounds of the Li_2O-MgO-Al_2O_3-SiO_2-CaO System—A First Survey. *Metals* **2020**, *10*, 1633. [CrossRef]
18. Wittkowski, A.; Schirmer, T.; Qiu, H.; Goldmann, D.; Fittschen, U.E.A. Speciation of Manganese in a Synthetic Recycling Slag Relevant for Lithium Recycling from Lithium-Ion Batteries. *Metals* **2021**, *11*, 188. [CrossRef]
19. Li, Y.; Yang, S.; Taskinen, P.; Chen, Y.; Tang, C.; Jokilaakso, A. Cleaner Recycling of Spent Lead-Acid Battery Paste and Co-Treatment of Pyrite Cinder via a Reductive Sulfur-Fixing Method for Valuable Metal Recovery and Sulfur Conservation. *Metals* **2019**, *9*, 911. [CrossRef]
20. Vieceli, N.; Reinhardt, N.; Ekberg, C.; Petranikova, M. Optimization of Manganese Recovery from a Solution Based on Lithium-Ion Batteries by Solvent Extraction with D2EHPA. *Metals* **2021**, *11*, 54. [CrossRef]
21. Klimko, J.; Oráč, D.; Miškufová, A.; Vonderstein, C.; Dertmann, C.; Sommerfeld, M.; Friedrich, B.; Havlík, T. A Combined Pyro- and Hydrometallurgical Approach to Recycle Pyrolyzed Lithium-Ion Battery Black Mass Part 2: Lithium Recovery from Li Enriched Slag—Thermodynamic Study, Kinetic Study, and Dry Digestion. *Metals* **2020**, *10*, 1558. [CrossRef]
22. Gerold, E.; Luidold, S.; Antrekowitsch, H. Selective Precipitation of Metal Oxalates from Lithium Ion Battery Leach Solutions. *Metals* **2020**, *10*, 1435. [CrossRef]
23. Doose, S.; Mayer, J.K.; Michalowski, P.; Kwade, A. Challenges in Ecofriendly Battery Recycling and Closed Material Cycles: A Perspective on Future Lithium Battery Generations. *Metals* **2021**, *11*, 291. [CrossRef]
24. Werner, D.; Peuker, U.A.; Mütze, T. Recycling Chain for Spent Lithium-Ion Batteries. *Metals* **2020**, *10*, 316. [CrossRef]
25. Brückner, L.; Frank, J.; Elwert, T. Industrial Recycling of Lithium-Ion Batteries—A Critical Review of Metallurgical Process Routes. *Metals* **2020**, *10*, 1107. [CrossRef]