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MECHANICAL PREPARATION OF END-OF-LIFE MILD HYBRID CAR BATTERY PACK

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Abstract: Lithium-ion batteries (LiBs) are essential to modern life, their uses range from small button cells to large-scale energy storage systems. While some used cells can be repurposed in energy storage applications, all lithium-ion batteries must undergo processing at the end of their life (EoL). The variety in shape, structure, and chemical composition of Li-ion batteries poses challenges for recycling. The first step in battery processing is mechanical preparation. This involves evaluating the structure of EoL lithium-ion batteries and designing an effective mechanical process. The process includes multiple comminution steps to liberate materials, separate parts, remove hazardous substances, and classify materials for Black Mass extraction.

Keywords: *lithium-ion batteries, mechanical processing, recycling, black mass*

1. INTRODUCTION

Recycling and processing lithium-ion batteries (LiBs) face several technical challenges and obstacles, including:

- **Variety of Battery Types**: The wide range of LiB types (such as LCO, LFP, LMO, NMC, and NCA) necessitates careful pre-classification and sorting to handle each type appropriately,
- **Size of Battery Packs**: The large size of battery packs and cells makes disassembly labor-intensive and challenging,
- **Flammability Risks**: LiBs are flammable and can ignite or explode due to mechanical stress or even improper storage,

- **Harmful Substances**: These batteries contain materials that are hazardous to human health and the environment, requiring careful separation, disposal, and recycling as part of a closed-loop or circular economy approach during mechanical preparation. Many types of Li-ion batteries (e.g., LCO, LFP, LMO, NMC, and NCA), require appropriate pre-classification and sorting of batteries.

The European Union currently has technologies for processing EoL LiBs, with capacities ranging from several hundred tons to 20,000 tons per year. Various methods, including pyrometallurgical, hydrometallurgical, and metallurgical techniques, have been suggested for recovering metals from LiBs (Roy et al., 2021). LiB processing technologies involving mechanical pretreatment are considered environmentally friendly, as they reduce the need for downstream operations to separate pure metals at later stages. Mechanical processes are generally cost-effective and do not typically require specialized equipment (Javorsky da Costa et al., 2015). In hydrometallurgical processes, mechanical processing serves as the first step, involving battery comminution, separation of structural materials, and extraction of the black mass (Brückner et al., 2020).

Implementing specialized processes can address several technical issues. For example, discharging batteries is often necessary before shredding to prevent short circuits and fire hazards (Mádainé et al., 2020). However, discharging may not be required with wet shredding or freezing methods. Shredding can also be performed in inert atmospheres (e.g., CO_2 or nitrogen) or under vacuum, and wet technologies are available for electrolyte removal, which can be achieved through thermal treatment or vacuum application (Greenwood et al., 2020).

The primary objective of this research is to extract the black mass from battery cells. A comprehensive material balance of the entire hybrid system and battery cell processing was conducted to use the results as a foundation for practical applications.

2. MATERIAL AND METHODS

2.1. Material

The experiments were carried out on Ford Transit mild hybrid car battery pack (Figure 1), from Auto Mandy Car Ltd., Budapest (Hungary). The battery package weighed 16.02 kg and included the battery cells, inverter, battery management system (BMS), connection circuits, and cooling system. The pack also included safety fuses and protective housing to prevent overheating and ensure operational safety. These components illustrate the intricate design of modern hybrid battery systems, emphasizing the challenges involved in disassembling, and processing these units.

Figure 1
Unassembled Ford Transit battery pack

2.2. Methods

The battery pack was manually dismantled using hand tools, which involved removing of the releasable connection. After this, the battery cells were discharged by connecting them directly to a resistance (Kwade and Diekman, 2018), followed by manual and mechanical processing.

The battery cell was opened using a two-axis rotary shear shredder. Once opened, the electrolyte solvent was removed by heat treatment (Colledani et al., 2023) using the Memmert UFE 400 type drying oven at 60 ℃ for 24 hours.

After the electrolyte was removed, the battery cell was mechanically crushed using a hammer crusher with a 20 mm sieve. The crushed battery cell material was sorted using an airflow separator with an air speed of 2.54 m/s. Separation was carried out into the following particle size fractions: >16; 16–12; 12–8; 8–4 mm. Particles smaller than 1 mm are referred to as black mass.

The qualitative analysis of materials and their identification was carried out on the FT/IR-4200 type A Fourier Transformed Infrared Spectrometer (JASCO) with a diamond ATR accessory. The spectra were obtained in a reflection mode.

The leached metals were analyzed using Varian Inc.'s 720-ES inductively coupled plasma (ICP) spectrometry. For the calibration of the device, a series of solutions were prepared from the certified multielement standard solutions, Certipur (distributed by Merck Ltd). and the Spectrascan (distributed by Teknolab).

3. RESULTS

3.1. The manual dismantling of the hybrid system

The material balance after dismantling is shown in Table 1. The system could be split into 5 units without destruction. The largest part was the battery pack (40.60 wt%). The battery pack was treated separately and is, therefore, not included in the table. This is followed by the system case, which consists of an iron base plate (2.64 kg) to which the hybrid system is attached, and a plastic cover (2.06 kg) held together by screws and weighs 5.29 kg. The air cooling fans, and the aluminium-copper heat exchanger of the inverter are integrated into the plastic cover. Next to it is the inverter, which consists of a large aluminium heat exchanger, an AC/DC converter, and control electronics. During disassembly, the removed screws, cables, connectors, and sensors were collected; together these weighed approx. 1.2 kg.

Table 1

Hybrid system units and their component materials (unit: wt%)

Material	Units name (wt%)			
	System case	Inverter	Cooling system	Other parts (screws, plugs, sensors)
Plastic	39.06	5.00	63.46	4.10
Iron	49.83	0.00	0.00	47.39
Aluminum	6.44	46.29	23.27	1.46
Glass	4.68	9.08	0.00	0.00
PCB	0.00	38.41	0.00	0.00
Rubber	0.00	0.00	3.24	7.23
Copper	0.00	1.21	10.04	24.04
Zinc	0.00	0.00	0.00	15.78
Unit/Battery system	33.38	17.13	1.22	7.67

3.2. Processing of the battery cells

During the research, not only was the mechanical processing of the battery cell carried out, but also the manual disassembly of a cell.

3.2.1. Results of manual dismantling

Manual disassembly of the battery cell structure and its quantitative and qualitative composition were investigated. In terms of construction, the battery is coated with a cellular composite casing, in which the anode, separator foil and cathode are layered.

According to the FT-IR measurements, the cell coating is a composite material consisting of polyethylene terephthalate-aluminium-polypropylene, PET-Al-PP. The cell casing provides 16.19 wt% of the total mass, with an aluminum content of 19–25 wt% based on volume concentration. The cathode foil consists of 28 aluminium plates, each 0.005 mm thick. It accounts for 2.51 wt% of the total cell mass. The anode foil consists of 29 copper plates, each 0.004 mm thick, making up 6.48 wt% of the cell. The separator foil between the anode and cathode plates provided 4.49 wt% of the total mass. Since the anode and cathode active materials could not be separated by the used technology, their mass fraction was determined as one, which amounted to 54.1 wt% of black mass.

During disassembly, the electrolyte content was determined by measuring the mass loss of the cells. The electrolyte content is 16.3 wt%, with a significant portion (10 wt%) removed shortly after opening the cell. The gas that is initially released contains mainly organic solvents (ethyl methyl carbonate [EMC]; dimethyl carbonate [DMC]) and CO_2 (Vetter et al., 2005; Aurbach et al., 2002).

3.2.2. Results of mechanical processing

Mechanical processing was carried out on electrically discharged batteries. The first stage of the process was to open the batteries and remove the electrolyte solvent with heat treatment. The 15.8 wt% electrolyte solvent left the cell, which is almost the same value as measured during manual dismantling. Since with this technology the electrolyte cannot be recovered, 100 wt% of the solid sample was used.

The particle size distribution and fractional composition of the battery cell after crushing in the hammer crusher are shown in Figure 2. According to the analysis, particles above 4 mm make up 60% of the sample's composition. Of these, 3/4 are metals with a partial black mass coating and 1/3 is separator foil. The fraction between 1 and 4 mm was composed of anode and cathode particles that were almost completely free of separator foil, with a significant coating of black mass. No further processing was done for particles between 1 and 4 mm.

(a) (b)

Figure 2

Main units of the hybrid system, (a) inverter and (b) battery pack.

The fraction below 1 mm, considered as a black mass, represented ~40 wt% of the sample. The composition of the extracted black mass, analyzed by ICP, is shown in Table 2. According to the results of the analysis, it has been determined that the battery that was processed is of NMC type. Upon analyzing the extracted black mass, it has been observed that it contains some amount of aluminum and copper foil fragments.

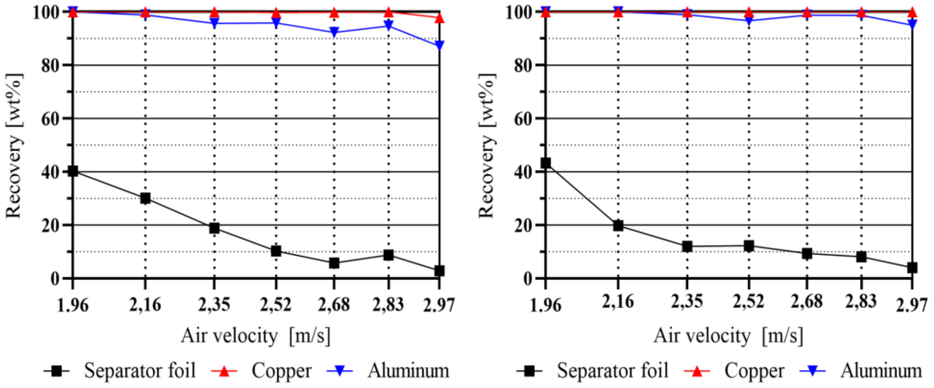
Table 2

Chemical composition of black mass

Chemical components	Ni	Mn	Cu	Ti	Al	Co	Fe	Li
wt%	17.3	3.49	3.95	0.98	0.91	4.53	0.07	3.39

3.2.3. Extraction of copper and aluminium

Valuable metals (aluminum and copper foil) with minor black mass coating were enriched in particle size fraction 4–16 mm. However, this fraction consisted of 70% separator foil by volume. The separation of metal and plastic was carried out in an airflow separator, with particle size fractions of 12–16- and 8–12-mm. Results of the separation are shown in Figure 3. Based on the measurement results, the optimal airflow velocity is 2.68 m/s. At this velocity, the metal content loss is at its minimum, but 95 wt% of the separator film will be transferred to the product with low settling velocity.



(a) *(b)*

Figure 3

Material recovery in the high settling velocity products in air separator by airspeed
(a) Particles between 4–8 mm; (b) Particles between 8–12 mm

4. DISCUSSION AND CONCLUSIONS

The cells can be well opened with a rotary shear crusher, after which the electrolyte can be completely removed by heat treatment at 60 °C for 48 hours. It was found that the technology used can recover approximately 74% of the black mass. Results show that more than 90% pure black mass can be obtained by mechanical processes. Further black mass is also found in the coating moulds of the product with a high settling velocity and on the surface of the 1 to 4 mm particles from the hammer crusher.

As the research progresses, we plan to classify the anode and cathode grains, and to remove and recover the black mass adhered to the surface of the particles.

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