Direct mechanical recycling concept for the recovery of black mass from battery electrodes

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Abstract. Ongoing climatic change, coupled with an increased awareness of sustainability and environmental protection, has led to the rapid development of renewable energy and power storage solutions. However, quality requirements and continuous technological development lead to a constant high level of waste in the production process, which is particularly critical in terms of resource efficiency. The scarcity of metals such as lithium, cobalt and nickel, which are widely used in LIBs, poses a major challenge to the European industry, in particular for the transformation of mobility and energy production. The industry already has established recycling approaches that break down valuable active materials into their raw materials, but have significant disadvantages in terms of resource consumption and emissions. Direct recycling is a newer, more sustainable recycling approach. It aims to recover the active materials of the battery cell using gentle and function-preserving processes so that they can be fed directly back into the production of LIBs. The key to direct recycling is the residue-free separation of the active materials from the other cell components. Following on from this, a mechatronic concept is being developed with which the active material of anodes is mechanically removed via brushing from the copper foil while preserving the morphological properties. The concept is then realised and tested in a test rig to demonstrate the functionality and suitability of the recycling concept.

Keywords: direct recycling, electrode, production scrap, recycling rate, battery

1. Introduction

The lithium-ion battery (LIB) technology has been continuously improved since the end of the 20th century and is now used in almost every mobile electronic device. In the 21st century in particular, research has made significant advances in the performance of rechargeable batteries. The scarce availability of metals such as lithium, cobalt and nickel, which are widely used in LIBs, poses a major challenge to European industry, in particular for the transformation of mobility and energy production. In terms of economic policy, a sustainable circular economy for battery technologies is firmly anchored in EU strategies. The EU Directive 2019/1020 requires recycling rates of 90% (95%) for cobalt (Co), copper (Cu) and nickel (Ni), and 50% (70%) for lithium (Li) by 2027 (2032) to ensure the recovery of strategic resources [1].

Although recycling focuses on the active material of the cathodes, recovering graphite from the anodes is also becoming increasingly important. It is becoming apparent that dependence on raw material imports poses increasing geopolitical risks [2]. In addition to the risks from the supply chains, immature processes also pose challenges for lithium-ion battery manufacturers. As a result, scrap quantities of up to 40 % are generated in the production process and should be recirculated immediately [3].

There are already two established recycling processes that each recycle lithium-ion batteries and their electrodes on an elementary basis. Hydrometallurgy relies on the dissolution, precipitation and filtering of the materials through chemical processes. This method requires high technical effort and large amounts of chemicals. Pyrometallurgy also relies on elemental decomposition. At high temperatures, electrodes and entire battery cells are melted down to recover the raw materials. Graphite and other organic materials are lost in the exhaust streams. The high energy consumption and climate-damaging emissions result in poor environmental performance. The functional properties of the active materials are completely lost in both processes. As a more cost-effective and environmentally friendly alternative, direct recycling of cathodes and anodes from used batteries and production scrap is being researched. In this recycling approach, the structure, function, and morphology of the electrode active material are to be fully regenerated, making it reusable [4].

So far, there has been no industrial implementation of the recycling concept due to the complexities of direct recycling [5,6]. The aim of this paper is to present a developed concept for direct mechanical recycling and to present and validate a physical demonstrator. The electrodes under consideration are single-sided coated anodes that have not yet been in contact with electrolyte. This paper considers the mechanical removal of the active material using brushes, with the aim of achieving high productivity and low solvent consumption in the recycling process. One challenge here is the purity of the active materials, which must not be compromised by the mechanical processing of the electrode. The aim is to ensure that no particles from either the current collector foil or the brush can be detected after the brushing process.

1.1. Direct recycling concept

The overall concept is a technological approach in the form of a process chain for the direct recycling of active materials (LFP and graphite) within the scope of the *DiRecFM* (WM34-42-57/28) research project, as shown in Figure 2. This project is dedicated to fundamentally improving the battery ecosystem. At the beginning of the process chain are coated and dried electrode sheets. These will first be delaminated in the process, and then the active material will be recovered through dispersion and centrifugation steps. The focus of this paper is on the development of the mechanical delamination concept for the electrode. Central to this is the residue-free separation of the active material from the current collector foil. This concept is to be subsequently realized and tested in a physical setup, which is also covered in this paper. For the proof of concept, the focus of this paper is on the recovery of the anode material, as shown schematically in Figure 2.

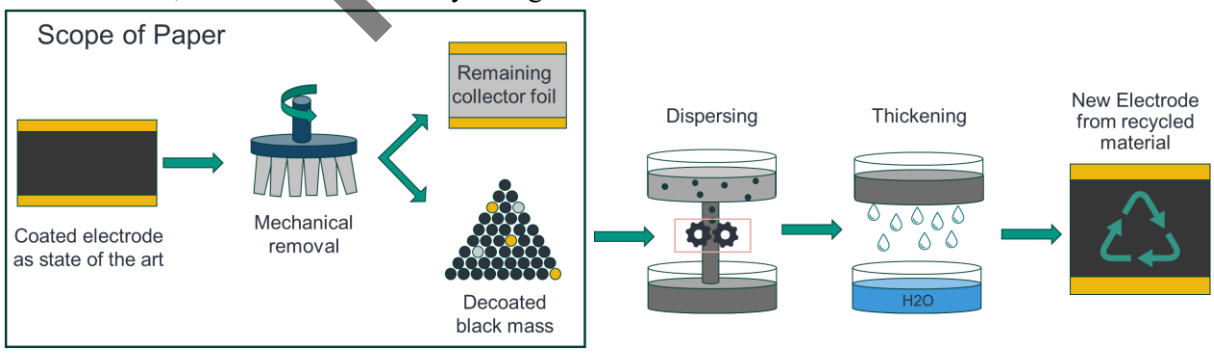

Figure 1. Schematic representation of the intended recycling process

2. Requirements for the direct recycling demonstrator

In a demonstrator that allows direct mechanical recycling by brushing and validates the concept, a mechanical and partially automated implementation is sought in order to enable high-quality and reproducible results. The following requirements for the test rig were identified:

1. Non-destructive, mechanical separation of active material and collector foil using brushes
2. Delamination of at least 95% of the active material
3. Avoidance of contamination of the active material by abrasion of the copper foil
4. Avoidance of contamination of the active material by the separation device
5. Processing of electrodes with dimensions of 250x200 mm
6. Forward movement of the electrode below the brush adjustable from 0.5 – 2 m/min

To address these requirements, different functions must be implemented in the demonstrator. The focus is on the separation of graphite and the collector foil by means of mechanical brushing. This must be realized in such a way that neither the copper foil is damaged nor the graphite is contaminated by copper or other materials from the brush. In addition to that and to achieve complete delamination, some kind of relative movement must take place between the brush and the electrode surface. In addition, certain setting options, such as relative speed and the force of the brush, should also be adjustable. Moreover, the anodes, which are available as individual sheets, must be fixed so that a complete treatment can be carried out. In order to protect the environment from the delaminated active materials, an enclosure must also be provided.

3. Development of the direct recycling demonstrator

The requirements were transformed into machine parts of the test rig using methodical design and construction according to VDI 2221 for product design and construction. The setup is divided into four functional units: the mechanical brush, a unit for performing relative movement, the electrode fixation, and the workspace enclosure to prevent the active materials from contaminating the environment.

3.1. Mechanical brushing

Preliminary experiments were executed to find a suitable brush material to mechanically delaminate the anode without the abrasion of particles from either the substrate foil or the brush. The electrodes were decoated by hand using the appropriate brush and adding water as solvent. The composition of the removed graphite was then analysed using an EDX analysis. Compared to materials such as brass and polyamide 6 brushes with larger filament diameters, polyamide 6 with a 0.3 mm diameter filament proved to be the most effective. [7].

For the realization of the relative movement between brush and electrode, a belt brush is selected. This solution ensures a uniform translational motion and thereby prevents local pressure loads and potential damage to the foil. Further, roll-to-roll processing is possible for the scale-up for recycling whole segments of production coils. The brush from *Wandres GmbH micro-cleaning*, as seen as a CAD model in Figure 2, is usually used for sensitive cleaning processes, for example lacquering or gluing surfaces. The continuous brushing, in addition to the movement of the anode, removes the graphite from the electrode surface and deposits it in the brush filaments. A unit for manual adjustment of the brush height via a handwheel enables different brush heights to be set to ensure various test modes. The graphite is then knocked off the brush at the deflection rollers of the brush belt by so-called knock-off pins. The addition of water to the electrode eases the decoating and additionally binds the active material in a slurry, preventing the rise of dust.

3.2. Forward conveyance and fixation of the electrode

The relative movement of the electrode and brush mechanism to ensure complete delamination is performed by a controlled linear axis driven by a stepper motor. This is controlled via an HMI control panel and can run at different speeds. With regard to the functions and dimensions of the linear axis, the

suitable slide size and the necessary lift are first derived based on the geometry and dimensions of the electrodes and their fixation. As reproducible test runs must be guaranteed for the validation of the overall concept, a repeat accuracy of +/- 0.5 mm is aimed. For an easy and fast fixation, a clamping concept is used for the interface between the electrode and the linear axis. Besides ensuring a defined position of the electrode, it is also important to not damage either the electrode or the brush by protruding parts or edges. Therefore, the electrode lies flat on a milled stainless plate and is clamped on both sides of the collector areas via screw-on steel plates as seen in Figure 3. The electrode surface is raised, so that the coating can be easily reached by the brush without any obstacles.

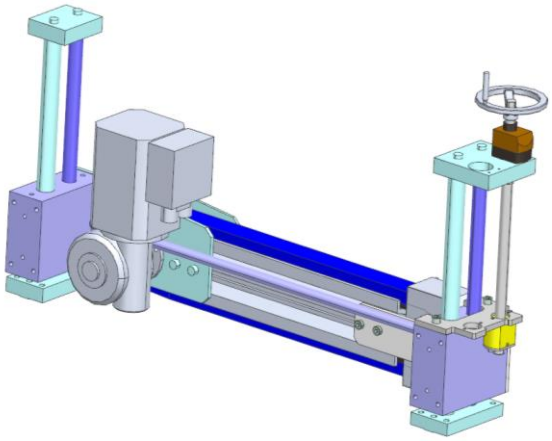

Figure 2. CAD model of the belt brush

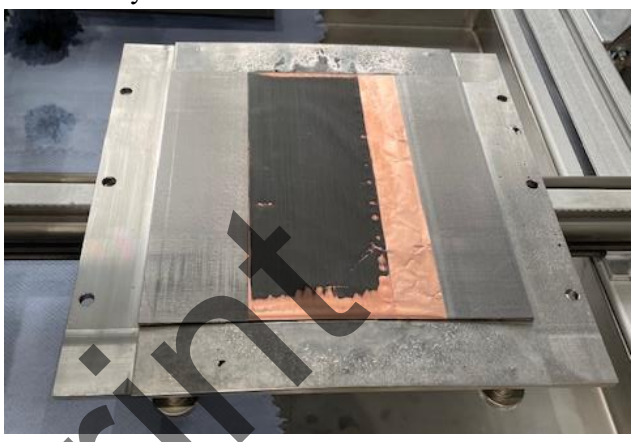

Figure 3. Electrode fixation

3.3. Enclosure of the demonstrator

To shield the plant operator and the immediate surroundings from the active materials and, at the same time, protect the removed active material from contamination, a suitable sealing concept is necessary. The binding of the active material particles by the water prevents the formation of dust, which tends to pose the greatest potential risk when inhaled or by penetrating electrical and mechanical components. The formation of aerosols of water and graphite is also a risk, as there is also the possibility that particles could be dispersed in the room and inhaled. For this reason, a complete enclosure of the working area is provided, as shown in Figure 4. Transparent polycarbonate panels are used to shield the experiments while maintaining visibility. In addition, the whole test rig is placed under an extraction hood to prevent any dust or aerosol released from escaping into the environment.

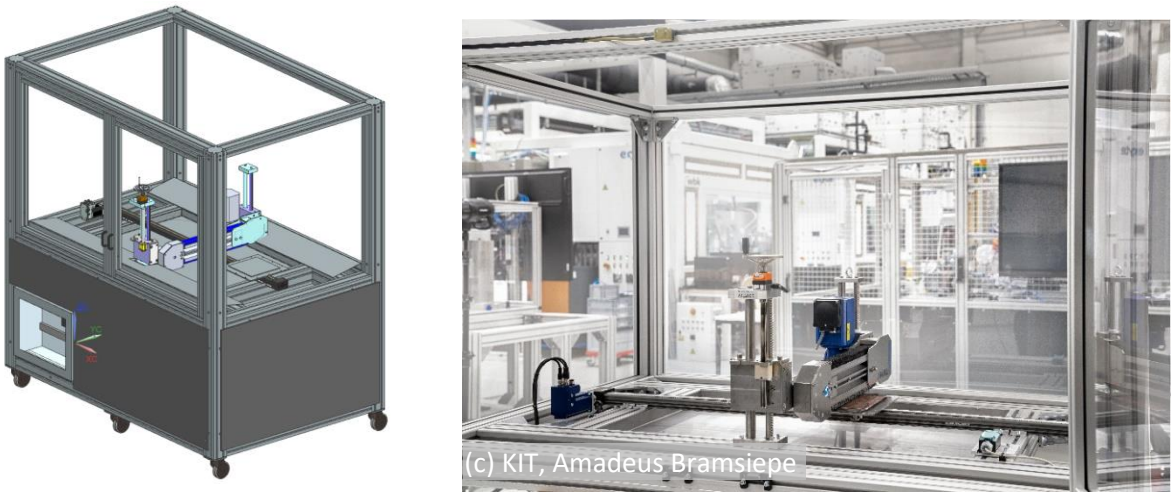

Figure 4. CAD model (left) and physical setup (right) of the demonstrator enclosure

4. Validation of the demonstrator concept

The procedure is standardized and consists of four steps as shown in Figure 5. After clamping the electrode on both sides (1), the surface is completely wetted with water (2). The actual decoating is then carried out using the belt brush (3) with the system closed. Subsequently, the decoated electrode active material is removed and analyzed (4). The parameters of the feed rate of the electrode and the brush indentation are varied manually between the tests.

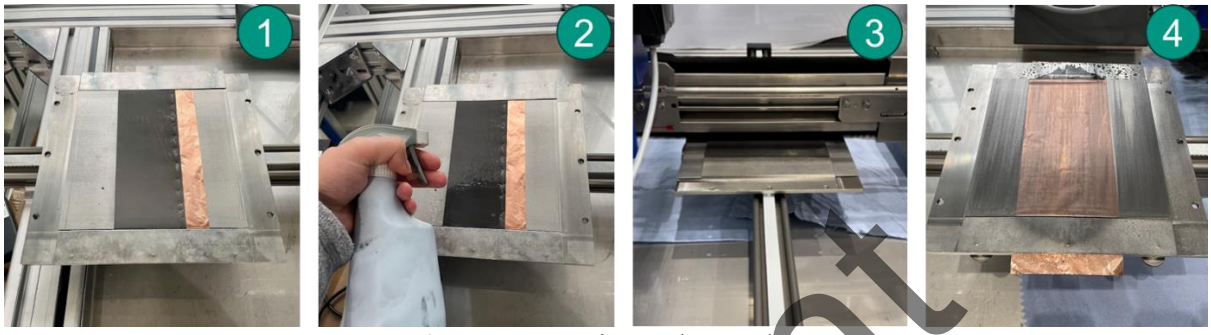

Figure 5. Experimental procedure

Of the identified requirements from Chapter 2, points 5. and 6. were fulfilled by the component selection. The contamination of the material (3. and 4.) has also already been checked in manual tests [7]. Therefore, the focus of the present study is on the freedom from damage and the high decoating rate of 95% to be achieved. A test plan is set up for the systematic investigation. The system parameters of the feed rate (v) and the brush indentation (δ) are varied as shown in Figure 6 (A) and (B). The former is varied between 0.5 m/min, 1 m/min and 2 m/min. The values 0 mm, 0.75 mm and 1.5 mm are set for the brush indentation. It should be noted that the brush indentation increases the pressure and bending of the filaments on the electrode surface. A full-factorial experiment is carried out in which a total of three repetitions are taken in the centre of the experimental design in order to be able to determine the repeatability of the results. The decoating rate and freedom from damage are analysed and documented. Every manual visible crack or plastic deformation of the collector foil is assessed as damage. For qualitative grading, cracks and irreversible deformations are rated with a 2. Any visible reversible deformation is rated with a 1. Only if there are no visible deformations or cracks is the evaluation result a 0. To analyse the decoating rate, the anode is weighed before and after the test using a precision scale and the measure of decoating is then calculated in relation to the theoretical weight per unit area of the coating.

The experimental design consists of a total of 11 variants and is a full factorial test in which each combination of the two variables is tested once. In addition, three tests are carried out at the centre point of the trials to detect any scatter in the results. Regarding the results, in terms of destruction of the electrode, it has been shown that no mechanical damage on a macroscopic scale, such as cracks or visible deformations, occurred during any of the tests. Even on the clamped copper foil, there are no recognisable traces of damage. Therefore, the damage can be rated with a 0 for each run.

The decoating rate, on the other hand, changes significantly by varying the feed rate and brush indentation. The aim of a decoating rate of 95 % is exceeded in a total of five runs. It can be seen that the lowest indentation of the filaments in runs 1 to 3 leads to inadequate results. As the feed rate increases, the decoating rate drops even further and reaches its lowest level in test 3. This result is also confirmed by the visual inspection of the electrode after decoating. In tests 4, 5.1, 5.2, 5.3 and 6, decoating rates of over 95% were already achieved in three out of five tests. An average value of 96.22 % was achieved in tests 5.1 to 5.3. The scatter shows a standard deviation of 3.12 %. Only trial 6

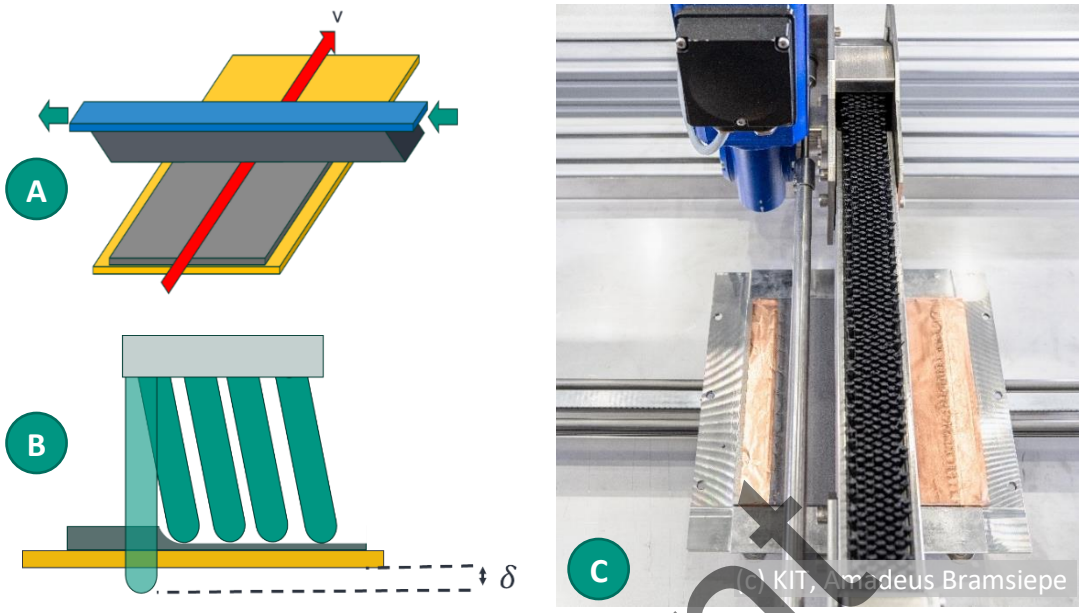

Figure 6. A: feed-thru speed v; **B:** brush indentation d; **C:** belt brush with partially decoated electrode

produced an unsatisfactory result of 62.54%. Here, too, it can be seen that a higher feed rate has a negative effect on the decoating rate. In the three test runs with the maximum indentation, very good results are consistently achieved at all feed rates. Test 7 achieves an almost complete decoating rate of 99.66 %. The unclean decoating that occurs in tests 1 to 6 with increasing feed rate is presumably compensated for here by the greater penetration depth of the filaments. The test results lead to two concrete statements: the penetration depth of the brush must be 0.75 mm in order to achieve sufficient results at feed speeds of 0.5 to 1 m/min. Furthermore, from a brush depth of 1.5 mm, the feed rate in the tested area plays a secondary role, as all speeds led to very good results here. The results are shown in Table 1.

Table 1. Results of decoating experiments

Run No.	Feed (m/min)	*Depth δ (mm)*	*Damage*	Decoating rate
1	0,5	0	0	18,56%
2	1	0	0	12,37%
3	2	0	0	1,37%
4	0,5	0,75	0	97,59%
5.1	1	0,75	0	92,10%
5.2	1	0,75	0	96,91%
5.3	1	0,75	0	99,66%
6	2	0,75	0	62,54%
7	0,5	1,5	0	99,66%
8	1	1,5	0	94,85%
9	2	1,5	0	97,59%

5. Conclusion and Outlook

The trials successfully demonstrate the feasibility of direct recycling using mechanical brushes for continuous processing. The direct recycling concept developed is therefore viable and suitable for the mechanical separation of active material and collector foil. Even though brushing off the electrodes' active material requires pre-treatment and complex material handling, it has advantages over other direct recycling concepts. The tests show that the feed rate is limited by the indentation depth of the brush filaments. However, it is conceivable to position the brush even deeper and increase the speed at the same time. The extent to which this could lead to damage to the collector foil or to contamination of the active material by particles from the collector foil must be investigated further. The processing of already damaged material, for example with partial web breaks, must also be taken into account at higher indentation. Another advantage of the developed concept is the economical use of solvents, as the interaction of the mechanical forces of the brush and the effect of the water led to very good results in the tests. This is to be investigated further in further tests by means of controlled feeding and dosing. Of interest here is the ideal application location and the smallest possible amount of liquid. The concept under consideration can therefore make an important contribution to the resource-saving production of LIBs by gently returning the active materials as recyclate to a second use cycle in the production.

6. Acknowledgements

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