



Recycling of low value components using high purity pre-treatment, direct recycling and green hydrometallurgical approaches for recycling of Lithium Ion and Sodium Ion batteries.

D1.2 Requirements for grading process

WP1 – Pre-processing, separation and delamination

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TABLE OF CONTENTS

1.	INTRODUCTION	4
2.	RECYCLING PATHS	4
2.1	GENERAL	4
2.2	MASS COMPOSITION OF LOW GRADE AND HIGH GRADE MATERIALS.....	4
2.3	MIXED FEEDS (CELLS, MODULES, BATTERIES)	7
2.3.1	PROCESS ROUTE 1: DRY SHREDDING MIXED FEED (COMMON APPROACH)	7
2.3.2	PROCESS ROUTE 3: DRY SHREDDING MIXED FEED (UoB).....	8
2.3.3	PROCESS ROUTE 3: WET SHREDDING MIXED FEED	8
3.	CONCLUSIONS	9

1. Introduction

Development of grading processes for active material into (1) high grade (as feed for direct recycling process) and (2) low grade (as feed for hydrometallurgical process). The target of developed grading process as 40% of output material is classified as high grade. This deliverable will be a result of Tasks 1.2 – 1.6.

2. Recycling Paths

2.1 General

The recycling path of direct recycling can only be applied to cathode active material and anode active material. Similarly, the alternative recycling path of hydrometallurgy is only relevant for cathode active material or black mass in general. Hence these materials should be graded, whereas other materials in LiB modules or cells that are part of the original input stream (aluminium foils, copper foils, separators, electrolytes, steel or plastic housings, etc.) are excluded from this definition.

For example, there are well-established metallurgic recycling routes for copper, steel and aluminium (up to a certain purity).

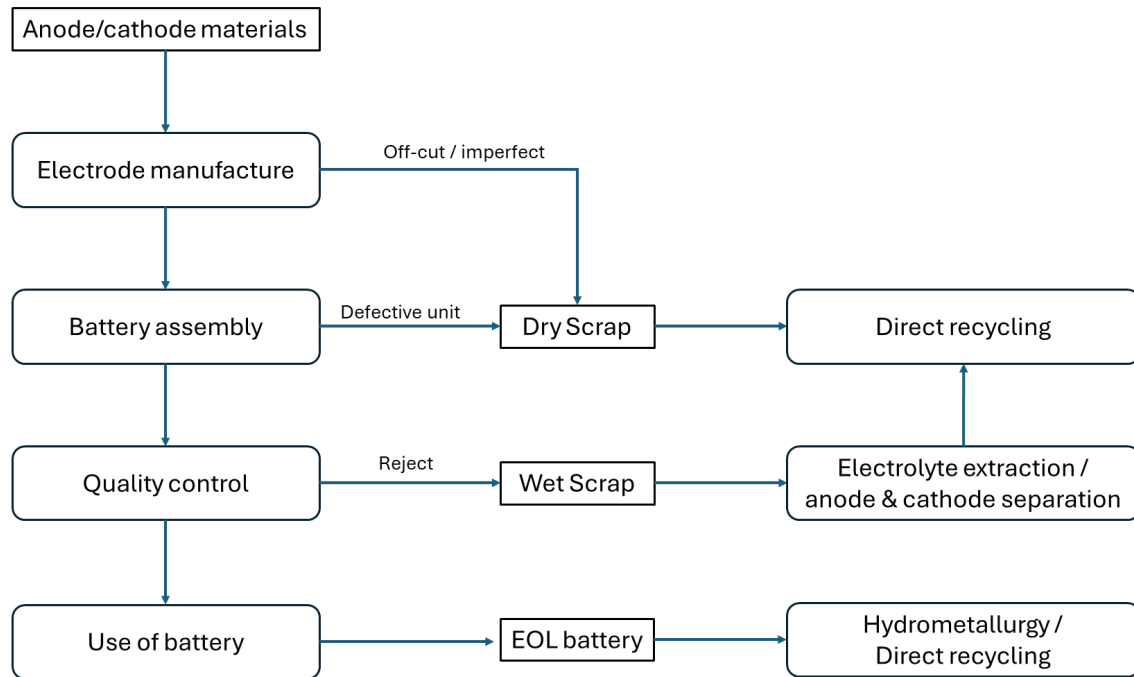
The grading depends on the process routes as well as on the material type (NMC; LFP).

2.2 Mass composition of low grade and high grade materials

The self-imposed goal within REVITALISE is to achieve 40% of material in a high grade that is reusable in direct recycling. Since black mass consists typically of about 55% to 65% cathode active material and 35 % to 45% anode active material, the 40% goal can be reached in various ways.

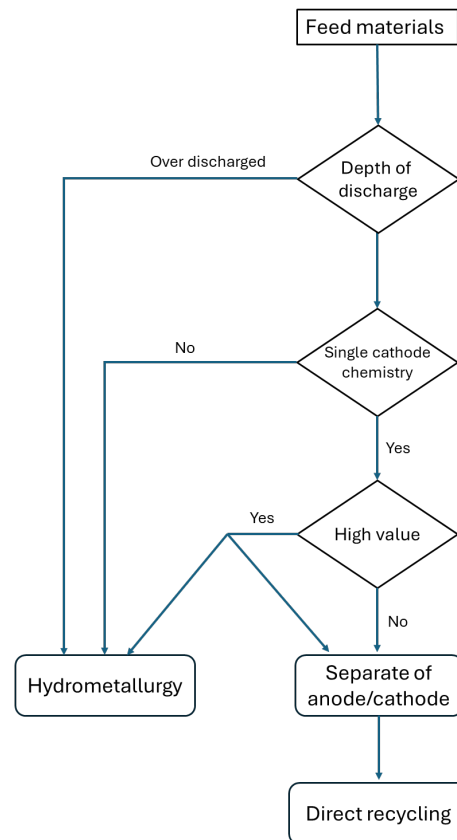
For example, the 40% could be reached entirely through cathode active material (perhaps possible for production scrap but unrealistic for batteries). More likely is that the goal is achieved through a combination of cathode and anode active materials, e.g. 20% of cathode active material and 20% of anode active material.

It depends on the recycling route, which portion stems from which type of material.



Firstly, there are two main sources of recycling materials, scrap coating and EOL batteries, which has different characteristics. It is important to understand that to apply different recycling strategies. The scrap coatings are generated during the manufacturing process and typically 5 – 30% of the total production depending on the maturity of the factory. The scrap coatings are further classified into dry and wet scrap by contact without or with the electrolyte. Dry scraps are from off cut and imperfect electrodes and defective unit from assembly process, and scrap active materials retains similar performance with pristine materials. Besides, the manufacturer knows the contents of scrap electrodes, and anode and cathode electrodes are well separated. Thus, dry scraps can be using direct recycling to recovery the active materials. Wet scraps are from Quality control rejections, and the anode and cathode electrodes are already packaged together. Hence the wet scraps need more steps to extract electrolyte and separate anode and cathode materials. However, for manufacturers, the content of the wet scraps remains defined and is still suitable for use in direct recycling.

Compared to scrap electrodes, the contents of EOL batteries are much complex. The EOL batteries contain not only solid-electrolyte interface, but also a cathode material in an uncertain lithiation state. Therefore, the materials of EOL batteries needs further grading process which is shown in next flow chart.



The first step is to judge whether the EOL batteries are over discharged or not. The over discharge causes irreversible damage of cathode structures, and the material need to be treated with hydrometallurgy. Subsequently, if the recycled material is a mixture of different types of cathodes (e.g. NMC mixed with LFP), it is hard to separate them, and they need to be sent to hydrometallurgy.

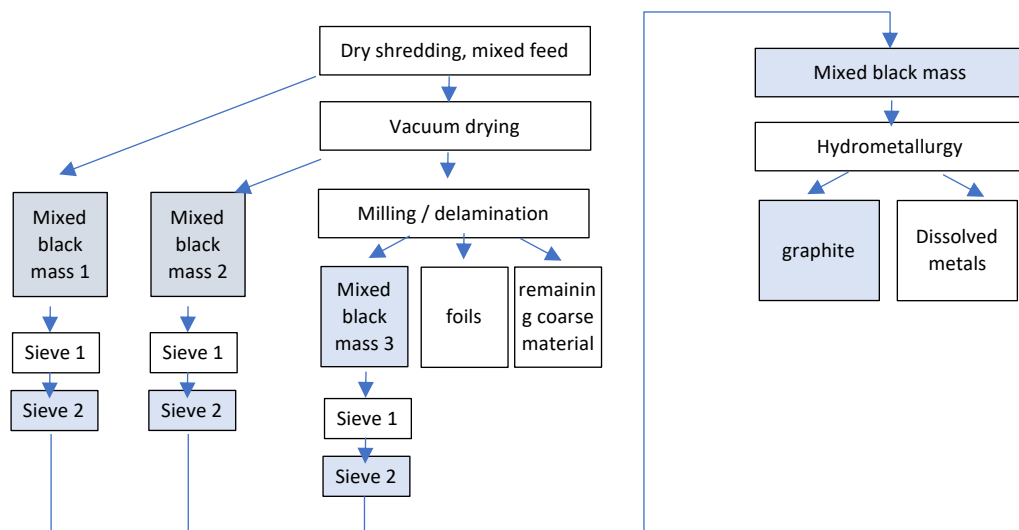
Then, the economics of recycling is considered. If the cathode materials have high value (such as LCO and NMC), it can be processed by either hydrometallurgy or direct recycling. For granular decisions on suitability of active materials for direct recycling, D2.1 includes more detail. LFP on the other hand has very low intrinsic material value, hence it ought to be recycled with direct recycling.

Furthermore, EoL batteries are likely to contain different amounts of active materials that are still suitable for direct recycling depending on their remaining state of health (SoH). Batteries with that retain a very high SoH (i.e. that had short usage life) are bound to contain more intact active materials that such with very low SoH. Whether SoH should be used to grade batteries remains to be seen depending on the outcomes of the project.

2.3 Mixed feeds (cells, modules, batteries)

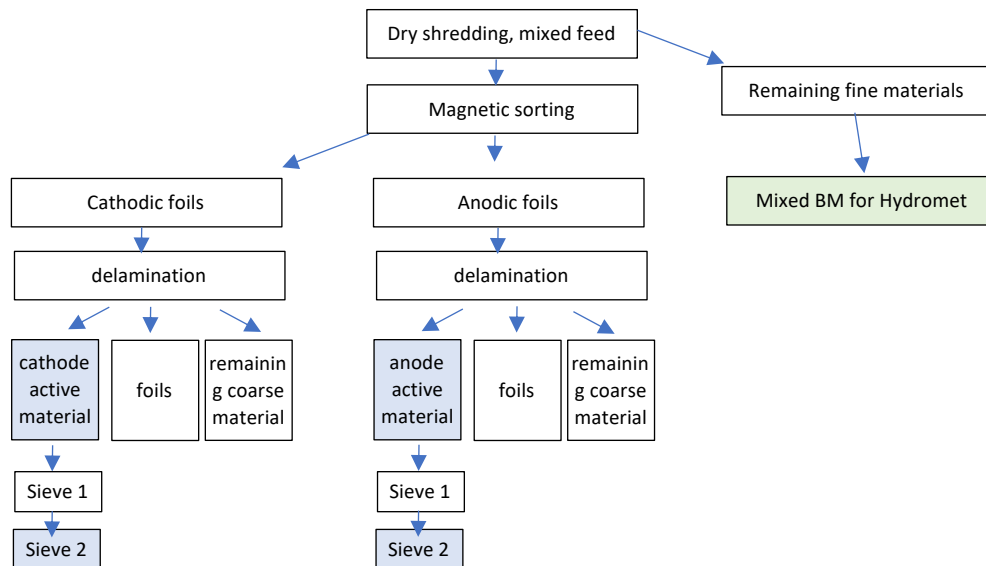
Assuming there is a feed of completely assembled cells, modules, or even batteries that are not over-discharged and not mixed with regards to chemistry (i.e. one type of cell only), then it is possible to retrieve some amount of the anode and/or cathode active material for direct recycling, depending on the pre-processing route.

2.3.1 Process route 1: dry shredding mixed feed (common approach)



The established dry-shredding process produces a mixed black mass which cannot be immediately used for direct recycling. However, a side product of the hydrometallurgical process is purified graphite. Depending on the hydrometallurgical processes, the surface of the graphite may be modified, requiring regraphitisation at 2000-3000 °C for extended periods. Materials which are intimately mixed through processes such as hammer milling are difficult to separate into high purity fractions suitable for direct recycling.

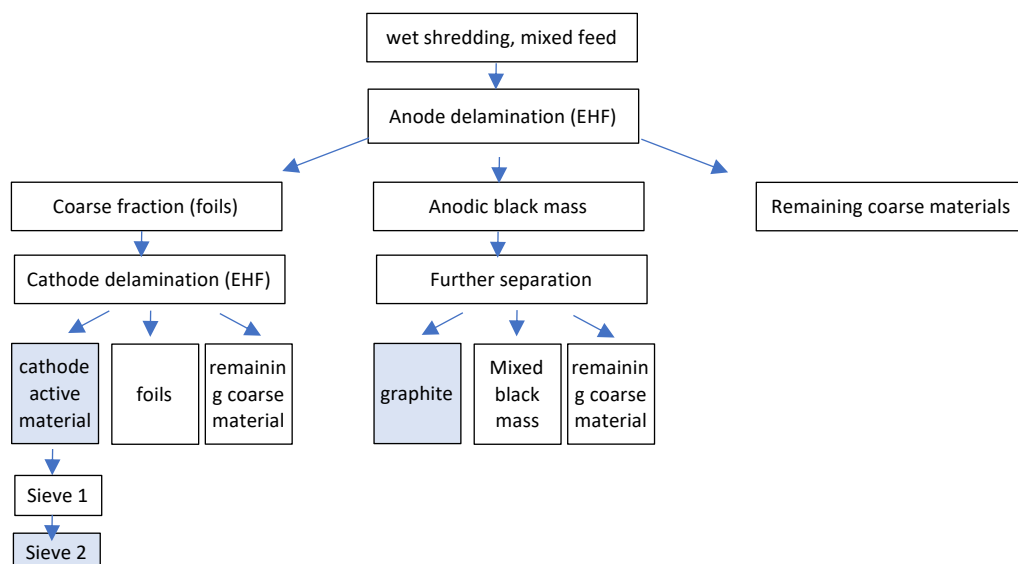
2.3.2 Process route 3: dry shredding mixed feed (UoB)



Ideally, the high grade and low grade fractions can be separated by a sieve.

In case of UoB's process route, certain amounts of both anode and cathode materials can be obtained that are principally suitable for direct recycling.

2.3.3 Process route 3: wet shredding mixed feed



In the wet recycling route, a part of the graphite and a large part of the cathode can be obtained at high grade qualities.

3. Conclusions

Single-component production scrap, such as cut-offs and imperfect rolls, can be processed separately and are high-grade materials. Dry scrap and quality control reject cells are also likely to be well suited for direct recycling, and should be considered high grade materials.

Direct recycling is best suited to single-chemistry feeds (e.g: just NMC622, with no LCO or LFP), of materials which have not been over-discharged. Black mass derived from mixed chemistries, over-discharged materials, or hammer-milled materials should be designated as lower grade materials suitable for hydrometallurgy as they are unlikely to be well suited to direct recycling. End of life cells, especially cells using obsolete chemistries are unlikely to be suitable for cost-effective direct recycling and should be designated for hydrometallurgy.

Single-chemistry black mass products, such as those where the active material separates at different points in the process, and is not intimately mixed through processes such as hammer milling are considered higher grade materials, and should be reserved for direct recycling.