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Influence of cell opening methods on organic solvent removal during pretreatment in lithium-ion battery recycling

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1 Introduction

Individual mobility, consumer electronics and electric energy storage are undergoing a technological transformation due to the invention of Lithium ion batteries (LIBs). At the moment, LIBs are applied already for most of the small- and medium-scale devices (Mossali et al. 2020), and contain critical, high value and important key engineering metals such as cobalt (Co), nickel (Ni), lithium (Li), copper (Cu) and aluminium (Al). The recycling of end of life (EOL) LIBs is promoted by legislation, mainly because of local environmental and health risks from hazardous materials, but also because of geostrategic and processing impacts as well as economic and supply chain effects (Harper et al. 2019, Mossali et al. 2020, Pinegar et al. 2019b, Rothermel et al. 2018).

However, the recycling of LIBs has also an environmental impact. In future, the production of LIBs has to be performed in closed loops. Therefore, battery recycling processes have to follow the developments in the LIB market and should be designed with the objective of compensating their life cycle environmental impacts by increasing the overall recycling efficiency (RE) (Kwade et al. 2018a).

One of the main challenges of LIB recycling is the batteries´ hazard potential and the corresponding depollution strategy. In this context, the depollution strategy significantly influences the RE, the process design for a safe battery cell opening and the energy demand of the whole process chain.

In order to evaluate and propose an overall disposal strategy regarding current and upcoming LIB applications, designs and compositions, the present investigation focuses on different methods for cell opening. The methods are combined with thermal drying to determine and compare the overall solvent evaporation or each cell opening method and dismantling depths. Consequently, technological and economical pre-treatment strategies for battery depollution and safe cell opening are discussed.

2 Recycling of EOL LIBs

2.1 Lithium-ion batteries

2.1.1 Design and composition

In principle, the functional unit of a LIB consists of a negative electrode (anode) of graphite or amorphous carbon compounds and a positive electrode (cathode) of a layered metal oxide. The layered oxide contains Li in combination with Ni, Co, Al and/or manganese (Mn) individually (LCO, LMO, LNO) or with different stoichiometry x, y and z on one hand ($N_xC_yA_z$, $N_x(M_y)C_z$) or iron phosphate (LFP) on the other (Zhao et al. 2019). These active materials are coated on an Al foil for the cathode and a Cu foil for the anode.

PVDF as well as carboxymethyl cellulose (CMC) combined with styrene-butadiene-rubber (SBR) are “state of the art” binders for cathodes and anodes, respectively (Korthauer 2019, Kwade et al. 2018b, Zhao et al. 2019). The binder acts as an adhesive for the coating material itself connecting it with the current collector foils. Carbon black is added as a conducting additive. Furthermore, petroleum coke, carbon fibre, pyrolysis carbon, glass carbon and carbon black may be added (Kwade et al. 2018b, Zhao et al. 2019).

A porous plastic foil, the so-called separator, separates both electrodes. The pores of electrodes and separator foil are filled with an ion-conducting electrolyte. The electrolyte is a high-purity multi-component mixture of organic solvents, conductive salt and further additives. Ethylene carbonate (EC), dimethyl carbonate (DMC), ethyl methyl carbonate (EMC), and diethyl carbonate (DEC) are the most commonly used organic solvents. DMC, EMC, and DEC are light boiling components with the boiling temperatures 90 °C, 107 °C and 127 °C respectively at ambient pressure. In contrast, the high boiling solvent EC boils at temperatures of 248 °C (Stehmann et al. 2018). Lithium hexafluorophosphate ($LiPF_6$) is almost exclusively used with as conductive salt in commercial LIBs (Yang et al. 2006).

The functional unit of electrodes and separator is piled or winded during cell assembling. As a result, the electrode-separator assembly forms stacks, or round as well as flat jelly rolls

(winding). The form of the functional unit determines the soft case pouch or hard case cylindrical and prismatic cell type, respectively (Korthauer 2019, Kwade et al. 2018b). Together, the functional unit and the hermetically sealed housing form the battery cell. Most of today's cells exhibit metal-based housing and packaging material. The housing contains further electrical connections, protection foils and functional components. Moreover, safety elements on cell level, such as burst membrane, overcharging protection components, and fuses, are added.

The cells as such are often connected in series or parallel. They form on the one hand a single block or on the other hand a module as subunit of a larger battery system (Korthauer 2013, Werner et al. 2020). Further peripheral functional and material components can be found on module or system level, such as battery management system, cooling, packaging, electronic and electric parts. As a general trend, all of the components are rising in complexity. Since cell manufacturers use their individual formulations (Zhao et al. 2019), the battery functional components, cells, modules as well as systems show a broad variety in used materials (Kwade et al. 2018b) and thus, overall material composition (Arnberger et al. 2012, Chen et al. 2019, Gaines et al. 2011, Georgi-Maschler et al. 2012, Kwade et al. 2018a, Kwade et al. 2018b, Mossali et al. 2020, Wang et al. 2016, Weyhe 2008, Wuschke 2018).

2.1.2 Hazard potential

All LIBs have hazard potentials due to their voltage and state of charge as well as due to their hazardous and reactive components. At the end of their lifetime, the hazard potentials of EOL-LIBs can be summarized as electrical, chemical, and thermal hazards (Rahimzei 2017). The hazard potentials interact with one another (Elwert et al. 2018, Gama 2014, Werner et al. 2020). Particularly, the components of the electrolyte require special care. The organic solvents potentially cause fire and explosion under special conditions (Hanisch et al. 2015, Wuschke 2018). Moreover, their hygroscopic properties promote corrosion (Kwade et al. 2018a). The conductive salt exhibits only limited chemical and thermal stability. During battery lifetime,

and especially under abuse conditions or in worst-case scenarios, flammable and toxic gases are generated as reaction or decomposition products. As a consequence of that, fire and several chemical reactions occur before, during and after battery cell liberation (Korthauer 2019, van Pels 2020).

2.2 Waste management

2.2.1 Complex waste

EOL-LIBs accumulate as unwanted production residues, but mainly as consumer residues either during or after the end of their use period. EOL-LIBs are future and highly complex waste with increasing complexity from the functional unit to the whole battery system (Pomberger et al. 2014, Rudolph 1999). Consequently, reverse production in terms of automated disassembly only could be realised at major expense by addressing the individual battery types and designs. The great inhomogeneity in structure and composition as well as the problematic ingredients of LIBs are challenging and require in high flexibility for disposal and recycling process design (Wegener et al. 2014).

2.2.2 Recycling chain and technologies

The recycling chain for LIBs consists of four process stages with two unit operations each (Martens et al. 2016, Werner et al. 2020). In the preparation stage (1), the batteries are usually collected (1.1) either separately or mixed according to the battery type. After collection, the batteries are sometimes if technologically possible sorted (1.2) with respect to either battery type (LIB, alkaline battery, Ni-metal hydride battery, lead-acid battery etc.), LIB chemistry (LCO, LMO, LFP, NMC), or even LIB active material ($N_1M_1C_1$, $N_6M_2C_2$, $N_8M_1C_1$). During the subsequent pretreatment (2), the batteries are dismantled (2.1) to defined dismantling depths. Also, the batteries are depolluted (2.2) regarding critical or hazardous components, or material conditions for the subsequent processes. The aim of the processing stage (3) is to liberate (3.1) the individual components or materials in order to separate (3.2) them physically

into defined concentrates. The refining of these concentrates occurs finally within the metallurgical treatment (4) using extraction (4.1) and recovery processes (4.2).

The RE of a process was established within recycling efficiency ordinance (EU) 493/2012 to quantify the usage of secondary (raw) materials for battery waste management and applied technologies. The RE is obtained by relating the cumulative mass of recovered secondary (raw) materials (output fractions) to the mass of batteries fed into the process (input/feed fractions) (Tytgat 2013).

The industrial recycling technologies for EOL-LIBs can be clustered to three process routes: low, medium, and high temperature route. The classification depends on the used temperature to depollute the batteries. The temperature influences the corresponding effort for preparation and processing, and overall RE (Werner et al. 2020). However, a high RE is reached only at the expense of special safety strategies for a secured process design. That includes the process medium and procedure for LIB system dismantling and liberation.

2.3 Dismantling

Disassembly of waste products conditions and reduces the feed material for further processing (Schwarz et al. 2018). Especially, functional components or reusable assemblies can be obtained for second-life applications (Harper et al. 2019, Idjis et al. 2013). Moreover, assemblies or components that can be fed to established recycling routes increase the overall RE (Elwert et al. 2018, Li et al. 2019, Wuschke et al. 2016). These materials decrease the complexity of down-stream liberation and separation as well as refining processes (Schwarz et al. 2018). The importance of dismantling strategies regarding their impact on the subsequent processing are rarely discussed for EOL-LIB recycling. Therefore, our contribution focusses on the interconnection between dismantling and depollution.

2.3.1 Methods

Dismantling is performed manually, semi-automatically (hybrid), or fully automatically (Elwert et al. 2018, Harper et al. 2019, Steinbild 2017). Manual disassembly is limited due to

economical and safety aspects. Hybrid concepts combine manual activities with industrial robots. Fully automated approaches use only industrial robots. The latter are in the focus of current interest and research (Ay et al. 2012, Harper et al. 2019, Treffer 2011, Zhao 2017a).

Generally, disassembly is an economic optimization problem between the dismantling depth, and the costs for the equipment and operating expenditure (Harper et al. 2019). Therein, the dismantling depth is a qualitative measure to describe the progress of disassembly in terms of generated parts, components, or its respective status (Nickel 1996). Typical dismantling depths are battery system, module, cell or electrode level. Additionally, the feed material for a dismantling step represents the lowest dismantling depth being increased by disassembly.

2.3.2 Opening of battery cells

Cell opening breaks up the battery cells´ housings to enable the separation of the individual components (Schubert 2002). It is part of the dismantling or liberation step (Werner et al. 2020) and presents an additional optimisation tasks of dismantling depth and mechanical and/or metallurgical processing (Marshall et al. 2020, Nickel 1996). The methods for cell opening are distinguished in manual or automatic procedures. Chipping in combination with disassembling is applied manually, whereas severing automatically. This contribution assigns chipping in combination with disassembling as manual cell opening within the unit operation dismantling. In contrast to that, automatic severing complies with mechanical cell opening similar to crushing/shredding and is therefore part of the unit operation liberation.

Chipping is done with geometrically determinate or indeterminate cutting edges in order to open the housing of battery cells. Metal and diamond saws, water or laser beams can be used for this purpose as well. However, using diamond saws and laser beams is very time consuming, lacks efficiency and causes fire hazard (Zhao 2017b). After chipping, the functional unit is removed manually. The following disassembly of the functional unit into its individual components is carried out manually as well.

2.3.3 Chipping in combination with disassembling

The combination of chipping with disassembling is known as “direct recycling route” (Chen et al. 2019), “full component recovery strategy” (Zhao 2017a) or „full component recycling process“ (Zhao et al. 2019). This approach is applied to recover the active material of the electrodes for the investigation of metallurgical, especially hydrometallurgical, refining treatment. This reverse engineering approach theoretically achieves the maximum RE. However, it is not industrially applied yet due to high personal effort and low throughput rates for consumer batteries in particular (c.f. 2.2.1). Altogether, specific procedures and processing times, especially for traction batteries, are rare in literature (Arnberger et al. 2018, Cerdas et al. 2018, Weyhe et al. 2016).

2.4 Depollution

The unit operation depollution prevents carry-over of critical or hazardous components or material conditions into subsequent process steps. In addition, depollution avoids the release of harmful emissions into the environment (Martens et al. 2016). The hazard potentials of EOL-LIBs are various: electrical, chemical, and thermal (cf. 2.1.2). Depending on the individual recycling process and its products, the depollution utilizes different methods, such as electrical, cryogenic, and/or thermal treatment, in order to remove hazardous substances or deactivate problematic conditions.

2.4.1 Methods and strategies

Electric and cryogenic treatment can be applied for the low temperature route. Electrical treatment includes mostly discharging to lower the remaining electrochemical potential of the battery (Harper et al. 2019). Therefore, discharging is essential for recycling processes which include dismantling to a high dismantling depth and/or mechanical liberation (Werner et al. 2020, Wuschke et al. 2019, Zhao 2017b). Cryogenic treatment avoids exothermic reactions due to the frozen and thus non-conductive electrolyte (Gama 2014). It prevents short circuits and

fires. Therein, the batteries are cooled via deep-freezing in liquid nitrogen (Kwade et al. 2018a, McLaughlin et al. 1999, Pinegar et al. 2019b).

Thermal treatment can be distinguished into pyrolysis and/or calcination. Those processes decompose the electrolyte components by breaking up the organic compounds thermochemically (Träger et al. 2015, Vezzini 2014). The energy released during this treatment is used as additional process heat. (Kwade et al. 2018a, Pinegar et al. 2019b, Werner et al. 2020).

2.4.2 Material flows

The evaluation of hazardous components as well as hazardous conditions depends strongly on the technological design of the individual recycling routes. Both, components and conditions influence the requirements and setup of the depollution strategy. Dismantling and depollution are carried out either downstream or iteratively. Moreover, electrical treatment is applied before thermal or cryogenic treatment from process technology aspects. Discharging is not necessarily required, so the latter can also stand-alone. Consequently, various combinations of procedures are possible, effecting the battery or material properties of the output streams (cf. Figure 1).

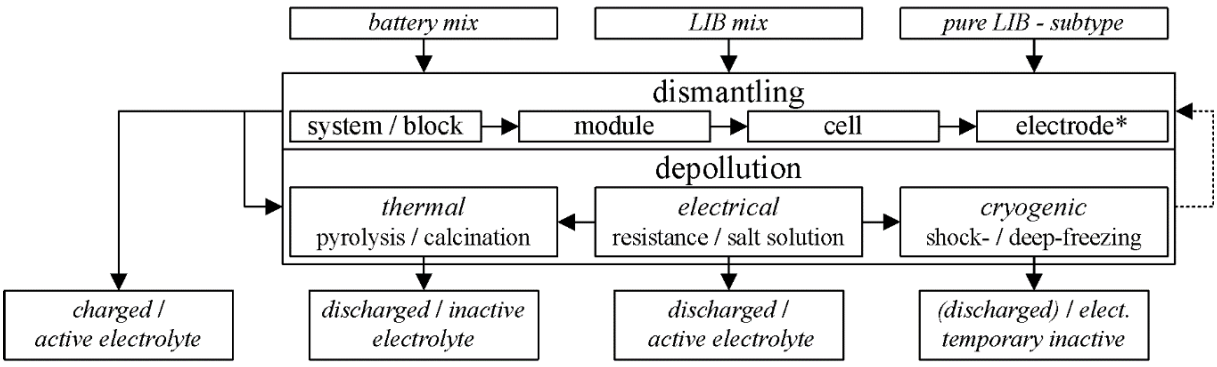

Figure 1 Dismantling and depollution as part of pre-treatment with potential in- and output streams, including respective material properties (* single electrodes are not charged (no electrical depollution necessary) but may still contain electrolyte (remaining chemical hazard potential)

The individual depollution strategy influences the subsequent separation processes. The depollution of the electrolyte is currently done before or after battery cell opening. After thermal treatment, the electrolyte is harmless in a subsequent cell opening (Weyhe 2008). In addition,

no special requirements are necessary for the process medium. Therefore, most of the industrial applications for LIB recycling use high temperatures to remove these pollutants. However, an appropriate exhaust gas treatment has to be added downstream and the theoretical RE is consequently reduced (Sojka 2020).

In contrast to thermal treatment before cell opening, various process designs remove the organic solvents and conducting salt after cell opening (Kwade et al. 2018a). If the electrolyte has to be recovered for material or energetic reuse, only temperatures below the decomposition temperatures of the plastics and electrolyte components are to be applied in the process design (low temperature route). Consequently, the highest theoretical RE can be reached only by three designs: discharging prior to cell opening (1), cell opening in ambient air with simultaneous solvent extraction (2) or using protective atmosphere with subsequent solvent separation (3) (Kwade et al. 2018a, Wuschke et al. 2015). Sojka (2020) presents a detailed but controversial and only qualitatively discussed overview of several process combinations with different depollution strategies and respective RE.

2.4.3 Process medium

For cell opening, the choice of process medium depends heavily on the hazard potential and depollution status of the feed material. An adequate process medium is often mentioned to design a safe cell opening process (Kwade et al. 2018a). This medium prevents easily and reliably explosive conditions with respect to lower and upper explosive limits (Stehmann et al. 2018). Therein, the high variation of feed composition remains a major challenge for a safe cell opening (cf. 2.1.2) accompanied by the differences in depollution status.

Besides aqueous media, other wet and also dry process media have been used to reduce the hazard potentials of LIBs during cell opening. On the one hand dry process media are used as protective gases such as argon (Fedjar et al. 2010, Valio 2017), carbon dioxide (Valio 2017), nitrogen (Steinbild 2017) or helium (Gama 2014). Besides of that, ambient air is used as process medium avoiding the explosions limits. The aim is to dilute and remove the liberated solvents.

Some of those applications operate at low air throughput using standard dedusting equipment. Other concepts use high throughputs generated by respective ventilators (Wuschke 2018). Finally, salt solutions are used as process medium (Valio 2017) containing calcium or magnesium (Woehrle et al. 2011).

3 Materials and methods

Manual cell opening and subsequent separation of organic solvents are examined and compared in this contribution. The temperatures for solvent separation during thermal depollution are selected in such a way that the plastics and electrolyte components are at maximum decomposed to a very small extent. Higher temperatures for battery depollution were examined by Weyhe (2008) as well as Weyhe et al. (2016). As a result, the influence of thermal depollution and dismantling depth on solvent release can be determined quantitatively and qualitatively.

The latter is seen as ideal procedure for component and subsequent material separation in comparison to mechanical liberation and separation. Consequently, the mass recovery of volatile organic solvents can be evaluated for the respective dismantling depths and manual cell opening method. Also, the distribution of solvents among the different solid components of a battery cell can be quantified. Hence, the method of manual cell opening allows an analytical determination of LIB composition and gives a reference of organic solvent vaporization during mechanical liberation.

3.1 Materials

A prismatic hard case LIB cell type (170 x 45 x 135 mm) is used due to its state-of-the-art status, current focus of interest, and its poor energy utilization, i.e. highest electrolyte content compared to the other cell types (Hettesheimer et al. 2017). The individual LIB cells origin from an automotive battery system consisting of 8 modules with 12 cells each (Weyhe et al. 2016). The burst membrane between the electrical poles of each cell shows a rectangular shape with 36 x 12 mm.

Table 1 shows the material composition of these battery cells determined by manual disassembly and separation. However, only qualitative data and the overall share for the solvents are available for the electrolyte since the amount of solvents was unknown and had to be determined by vacuum drying. The solvent consisted of DMC, EMC and DEC as highly volatile components, and EC as lowly volatile ingredient. The conductive salt and additives remain theoretically within the pores of the other cell components.

component	**function**	**material**	***w* in %**
cathode	metal foil	aluminium	3.0
	coating	NMC + PVDF + additives	34.0
anode	metal foil	copper	7.2
	coating	graphite + SBR + CMC + additives	17.9
housing	NSD contact	copper	0.9
	electrical contact	copper	0.7
	electrical contact	aluminium	0.3
	case	aluminium	11.8
	retainer	PP	0.7
	sleeve	PET	0.3
	NSD foil	PP	0.1
	foils	PP	0.2
	glue		0.2
	others		0.5
separator	foil	PP/PE/PP	1.9
electrolyte	organic solvents	DMC EMC DEC EC	16.8
	conductive salt	LiPF6	2.6
	additives		1.0

Table 1 material composition of the used battery cells

3.2 Experiments

Complex and multistep experiments in laboratory scale were employed, combining dismantling and depollution (cf. Figure 2). Dismantling of a battery system (*system*) to module and cell level was carried out via manual disassembling, using several tools to liberate the different components. Electrical depollution was performed at module level with an electrical resistance. Afterwards, the cells were opened manually in normal air. The burst membrane was grooved with a common slotted screwdriver (Figure 2: *open membrane*). The cell housing was opened

with a 300 mm hacksaw cutting the three smallest side panels of the prismatic cells, which did not contain the electrical poles. Then, the housing was bent open and the electrical contacts were removed to detach four windings (c.f. Figure 3 left) using a common pipe wrench.

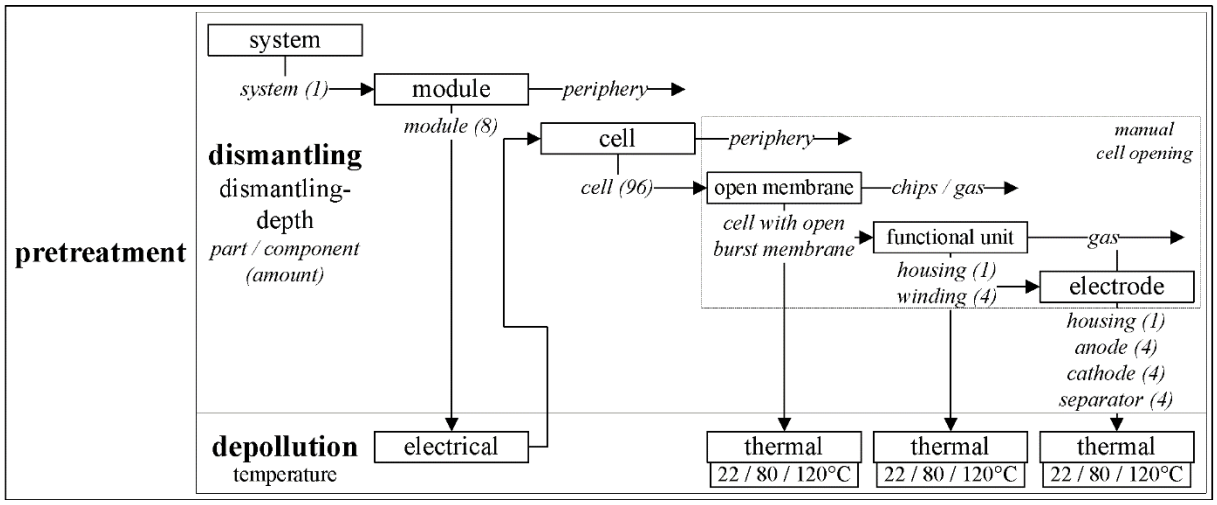

Figure 2 Flow chart of experimental procedure

The protection foils were manually pulled of each winding (Figure 2: *functional unit*) and were added to the housing components (Figure 2: *housing*; c.f. Figure 3 middle). Finally, the windings were unrolled and *anode*, *cathode* and *separator* were separated manually as well (c.f. Figure 3 right). The mass of the individual components as well as the time for each dismantling step were recorded.

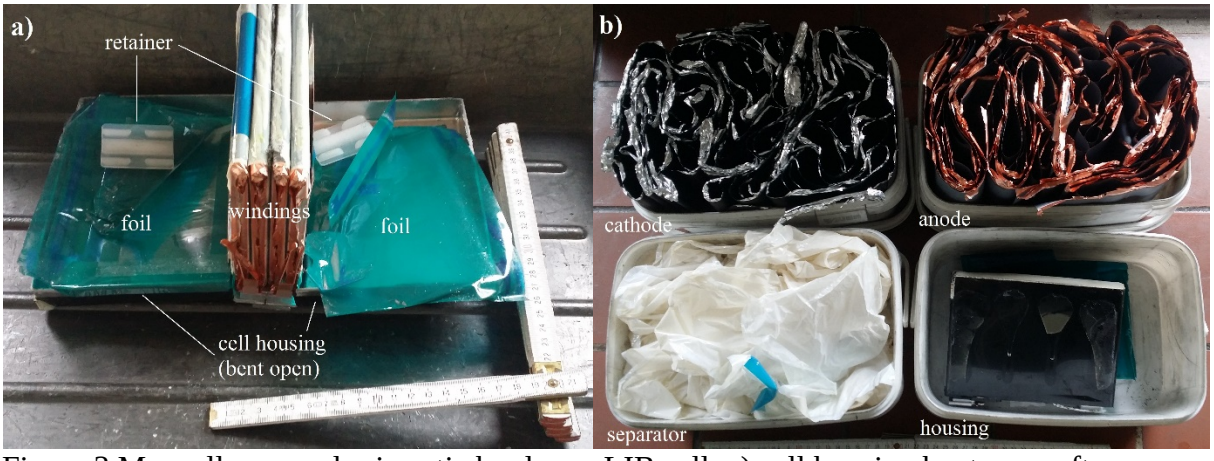

Figure 3 Manually opened prismatic hard case LIB cell: a) cell housing bent open after chipping; b) separated cell components with empty cell housing in closed position and unrolled components of functional unit (four windings from a))

The organic solvents were evaporated for cells with open valve, the functional units and the electrodes via thermal drying under ambient atmosphere. Therefore, a laboratory fume under

283 room temperature (ca. 22 °C) and two laboratory drying chambers (HERAEUS t 6420 for 80 °C
284 and MEMMERT universal oven UF 110 for 120 °C) were used. Three temperatures (22 °C,
285 80 °C, 120 °C) and two drying times (1 h, 120h) were set for solvent evaporation representing
286 the immediate as well as a long term release. The selection is based on studies in literature (He
287 et al. 2015, Pinegar et al. 2019a, Stehmann et al. 2018, Wang et al. 2016, Zhang et al. 2015)
288 and the melting temperature of the separators´ plastics, which should not be exceeded. The
289 numbers in brackets in Figure 2 represent the amount of parts or cells disassembled from *system*
290 to *cell* level. The ones for the dismantling depth *functional unit* and *electrodes* represents the
291 components after the *cell* was manually opened. Three cells were investigated for each
292 depollution temperature of the respective dismantling depth. Thus, in total 27 cells were
293 prepared for the experiments.

294 3.3 Methods

295 The processes of dismantling and thermal drying are analysed by the mass balance in respect
296 to the influence of different pre-treatment strategies. Therefore, the in- and output mass m_i and
297 m_o of individual materials (cell, cell with burst membrane open and winding) and components
298 (housing, anode, cathode, and separator) as well as the time spent for the respective process
299 step was determined. With that, the throughput of manual dismantling $\dot{m}$ could be determined
300 as relation of feed mass and processing time. If several dismantling steps were carried out, the
301 output mass of the first step equals the feed mass for the following one.

$$\dot{m} = m_i / t_r \quad (1)$$

302 The mass difference between in- and output mass is associated either to solid parts of the battery
303 system and module periphery or gases from evaporating organic solvents or their respective
304 decomposition products. The amount of dust or small fragments like chips is negligible. The
305 relative mass difference Δw is related to the cell mass representing in general only evaporated
306 solvents.

$$\Delta w = (m_i - m_o)/m_{cell} \quad (2)$$

In contrast, the relative amount w_{os} of organic solvent for one of the cells components *j* (housing, anode, cathode, and separator) is calculated by the evaporated mass of each component *j* in relation to the overall evaporated mass of solvents at the respective temperature.

$$w_{os,j} = (m_{i,j} - m_{o,j})/(m_i - m_o) \quad (3)$$

The mass reduction of solvents equals the recovery of solvents in the theoretical solvent fraction $\Delta w_{os,j}$. This fraction represents the total amount of solvents evaporated from a component and is determined by the product of mass recovery R_m at the respective temperature and each component´s mass reduction of organic solvents $w_{os,j}$ in relation to the cell´s organic solvent content $w_{os,cell}$.

$$R_m = m_{material\ fraction}/\ m_i \quad (4)$$

$$\Delta w_{os,j} = R_{os,j} = R_m \bullet w_{os,j}\ /w_{os,cell} \quad (5)$$

Often, the mean value (MV) as well as minimum (MIN) and maximum (MAX) will be highlighted for a better overview.

4 Results and discussion

4.1 Dismantling and manual cell opening

Each dismantling step is accompanied by a certain decrease of remaining mass m_0 due to removal of solid and gas components (cf. Figure 4 left). The solid components consist of system and module peripheral parts like system and module housing, battery management system, thermal regulation system, electronics and electrical wires and connectors. These components can be fed into already established recycling routes benefiting the recycling efficiency of the battery itself. The gases correspond to the evaporated organic solvents contained in the electrolyte, which are volatilized during cell opening or its decomposition products. Process time and dismantling costs increase with increasing dismantling depth since more components and compounds have to be treated individually. Especially, manual chipping and unrolling the

electrode stacks are time-consuming processes (Wuschke 2018). As a result, the throughput of such a process decreases with increasing dismantling depth of manual cell opening (cf. Figure 4 right).

The disassembly of battery systems to modules or cells generates only solid components. The share of chips generated by hacksawing the cell housing is negligible (< 0.01 %). Comparing the different dismantling depths, disassembling to electrode level shows the maximum possible mass reduction for subsequent mechanical processing and the most efficient evaporation of the solvent due to tremendously enlarged surface area.

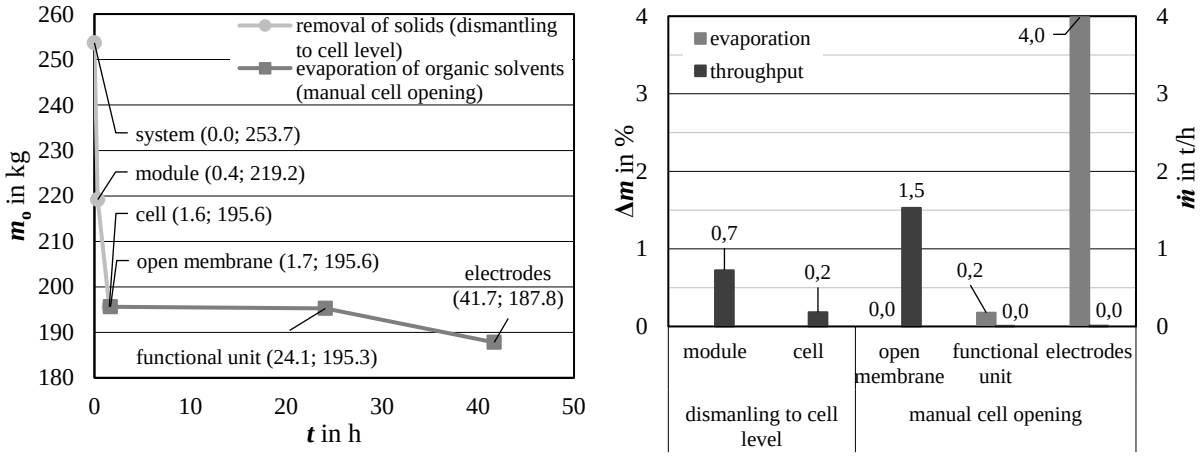

Figure 4 left) output mass and process time for different dismantling steps; right) influence of process step on mass reduction by evaporation and on possible throughput for the respective dismantling step

From a design point of view, the individual windings of the battery type investigated are almost completely enclosed by an additional plastic foil (c.f. Figure 3 left and middle). With that, the electrolyte cannot be released from the pores of the coating materials and the separator during dismantling. Only small amounts of solvent evaporate at ambient conditions if the burst membrane or the housing is opened. In contrary to that, dismantling to electrodes level increases the volatilization of solvents. That may be equated with the uncontrolled stressing by arbitrary tools, which damages usually the functional unit or liberates partially or completely its components.

Solvent removal represents the reverse process of electrolyte filling within cell production. During cell production, electrolyte filling is the most time-consuming sub-process. Therefore, pressure and temperature are already optimized in this field (Knoche et al. 2016). Consequently within recycling, solvent removal takes quite long as well. Most of the solvents appear to be contained inside of the pores and only a small proportion on the outside of the plastic foil.

In principle, the evaporation of solvents increases with increasing dismantling depth. As soon as the functional unit is partially liberated, more components and more mass of solvent volatilize due to a larger free or active surface area. Especially low boiling solvents like DMC, DEC and EMC are emitted in the initial steps generating a hazardous, i.e. explosion potential (Stehmann et al. 2018). Therefore, cell opening has to comply with the explosion limits of such a mixture to provide safe operation conditions.

4.2 Depollution

Thermal depollution of the LIBs at different dismantling depths was examined for three temperatures and two drying times. Naturally, the mass due to evaporation increases with higher temperature and longer drying times. If the previous dismantling is taken into account, the cumulative mass difference depends also on the surface area (cf. Figure 5). Therefore, the functional unit and especially the components on electrodes level show a faster drying kinetic compared to the case of the opened burst membrane. Within the first hour of drying, a big share of solvents is already evaporated, even increasing with higher temperatures.

Cells with open burst membrane and the functional units show the biggest effect of evaporation between 1 and 120 hours due to different heat and mass transfer through the materials. As a result, only high surface area, long drying time and high temperatures separate the volatile electrolyte components to a sufficient degree. Otherwise, especially the organic solvents with high boiling point remain in the cell or cell components. Stehmann et al. (2018) stated that if the temperature is the main driver for solvent removal via thermal drying, information on the boiling temperature of the solvents contained is required to estimate the drying potential. Since

the electrolyte is a solvent mixture, the relative volatility of the organic components determines the drying efficiency. Especially the low volatile solvents tend to remain with the solids, either adsorbed or still as liquid phase within the porous structure of the materials. It is supposed that with increasing drying temperature the solvent removal becomes more and more complete. In order to separate the low volatile solvents, the drying temperature has to be adjusted or rather increased (Sattler 2001, Stehmann et al. 2018).

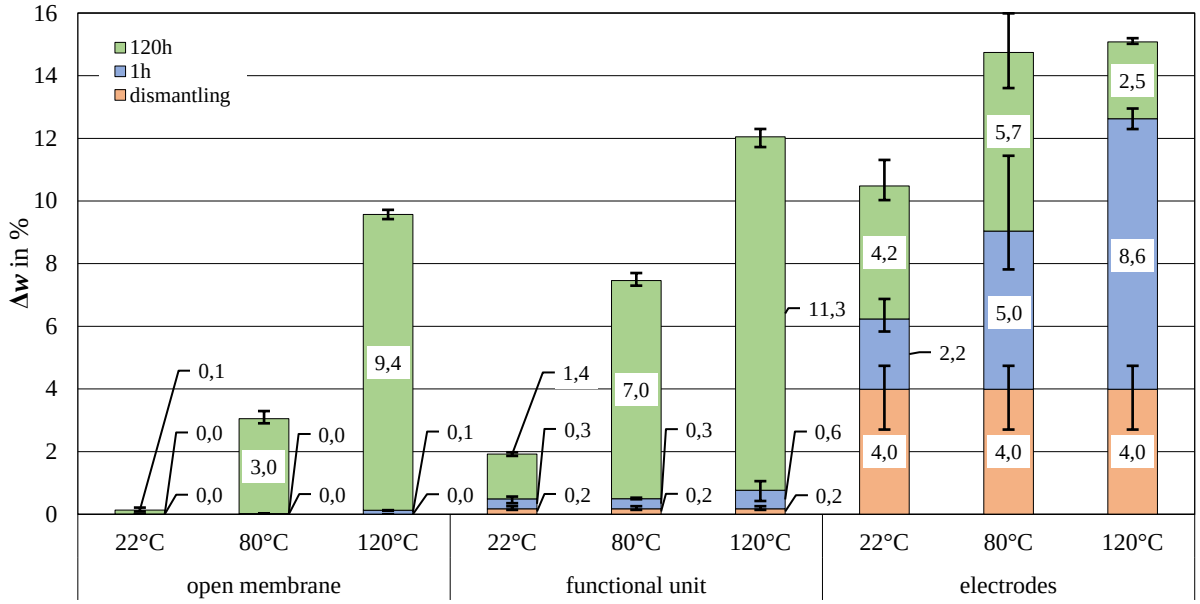

Figure 5 Influence of drying temperature and time on mass difference for different dismantling depths

Figure 6 shows the cumulative reduction of organic solvent over all steps during thermal depollution after 120 hours drying time for the individual cell components as well as the overall pretreatment of electrodes, consisting of the process chain from manual cell opening to depollution (cf. 3.2). The majority of the organic solvents are on the surface as well as in the pores of the components. The housing and its plastic foils show only small amounts of superficial solvents, so the corresponding mass reduction is hardly measurable and negligible. Irrespective to the temperature used, the mass reduction of organic solvents increases with increasing material thickness and pore volume of the components. With respect to the organic solvent content measured at electrode level, more than 90 % of solvents are already removed

during dismantling and drying at 120 °C. The relative amount of organic solvents evaporating from the individual cell components is independent on the temperature (cf. Table 2).

The results of thermally treating the components at electrodes level at 22 °C indicate that around 10 % of the cell representing 63 % of the total solvent content consists of low boiling components DMC, EMC and DEC. If the temperatures are raised, the evaporation kinetics of the low boiling solvents is increasing and, in addition, the high boiling components start to evaporate, even the total relative mass difference between 80 °C and 120 °C at 120°h remains equal. However, the used temperatures or residence times are not sufficient in order to evaporate the high boiling solvent EC, which has to make up the other 37 % of the total solvent content. According to Yang et al. (2006), no conductive salt decomposes, as long as solvents are in presence. Hence, no hydrofluoric acid (HF) is created during the used thermal treatment setup.

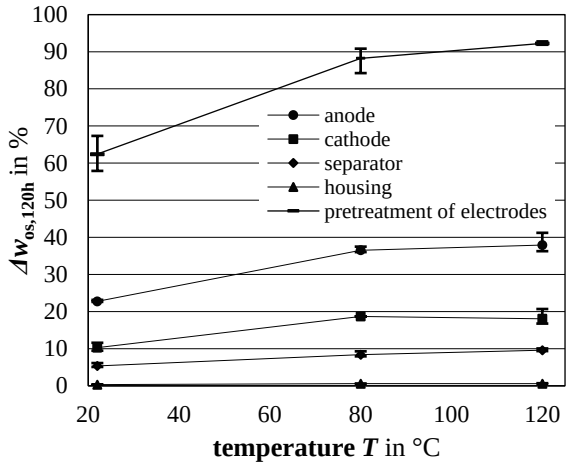

Figure 6 Influence of drying temperature on reduction of organic solvents with respect to amount of organic solvents in the component (MV; MIN; MAX)

component	temperature T in °C	organic solvent share $w_{os,120h}$ in %		
		MV	MIN	MAX
anode	22°C	58,8	58,4	59,4
	80°C	56,9	56,1	57,7
	120°C	57,3	54,8	59,8
cathode	22°C	26,6	24,4	27,8
	80°C	29,1	29,1	29,1
	120°C	27,3	25,3	29,3
separator	22°C	13,8	13,1	15,1
	80°C	13,1	12,4	13,8
	120°C	14,5	14,1	14,9
housing	22°C	0,9	0,7	1,0
	80°C	0,9	0,8	1,0
	120°C	0,9	0,7	1,0

Table 2 Influence of drying temperature on the share of organic solvents in the individual cell components measured at electrode dismantling level after 120 h drying

4.3 Proposal of a recycling chain

Dismantling LIBs to functional unit or electrodes level simplifies or even dispenses further sorting steps due to the removal of the housing components. Unfortunately, the necessary manual dismantling consumes time and labour force, which makes it economically unfeasible. Therefore, automated dismantling or mechanical processing incorporating crushing is the most promising approach. Both of them have to adopt the challenges coming from the high variety

of LIB (c.f. 2.2.1 and 2.3). Their complexity in design and material compositions increases with the ever-growing variety of applications and special requirements of LIB, especially for automotive applications with respect to fast charging and high range. The variety of cell compounds differs from modular and easily demountable to agglutinated throwaway types with a multitude of different dimensions and designs.

Thermal depollution of LIBs with open burst membranes can theoretically skip discharging and avoids a protective atmosphere during cell opening (Pinegar et al. 2020). This results in a less complex design of process technology as well as economic benefits regarding capital and operational expenditure. However, this approach shows disadvantages like a long process time and insufficient solvent removal. The necessary energetic input and expenditure of time for solvent removal does not outweigh the benefit of a simplified subsequent separation of the remaining solid materials and an increased recycling efficiency. Furthermore, pre-sorting is necessary regarding cells with an easily accessible and openable burst membrane. Opening cells without a burst membrane by methods like drilling or sawing gives access to the functional units, but require additional safety measures. In this case, solvent removal will be similar to the methods described in this investigation. As a consequence, only temperatures at or above 250 °C present a feasible option as an adequate depollution strategy for all cell types. The so called moderate temperature route causes cell disruption by the increased inner pressure of the cell during thermal depollution, but is only accompanied by a limited recycling efficiency (Sojka 2020, Weyhe et al. 2016).

The amount of removed solvents increase with the dismantling depth due to higher surface are for thermal depolluted functional units or electrodes. It therefore remains doubtful from a thermodynamic perspective whether all solvents can be separated at ambient conditions, as especially high boiling solvents require higher temperatures or lower pressure in order to evaporate. Regarding process designs, if recycling technologies avoid a drying step or apply a drying step only at ambient conditions after cell opening, they have to deal with further

continuous solvent evaporation in the subsequent physical separation or metallurgical refining processes. The solvents will basically contaminate all generated material fractions, whereby the housing fraction only in small amounts. Moreover, processing wet materials is not desirable (Diekmann et al. 2018), and enclosure of the processing equipment is expensive and complicates maintenance work.

5 Conclusion

LIBs have several hazard potentials, which have to be deactivated for the save processing of EOL material. The hazard potentials originating from the organic solvents of EOL-LIB are currently deactivated cryogenically or with high temperatures before cell opening. The latter is applied in particular on industrial scale at the high or the moderate temperature route. Alternatively at low temperature routes, the organic solvents have to be separated after cell opening. The interdependence of dismantling and depollution during the pretreatment of EOL-LIB have been examined in order to determine their influence on solvent extraction. Within this scope, different recycling chains have been tested and discussed.

In general, the evaporation of organic solvents increases with increasing dismantling depth, temperature and time for (thermal) drying. Dismantling to electrode level evaporates already around 25 mass percentage of the contained solvents increasing the drying kinetics due to higher surface area. The mass loss can be mostly correlated to the evaporation of the low volatile organic solvents. Also, the highest share of solvents evaporates directly from the surface and the pores of the anode, followed by the pores of the cathode and separator. Within that, the temperature does not influence the relative amount of evaporated solvents. Therefore, the evaporation kinetics are influences more by morphological features of the cell components instead of the drying temperature and regime.

The investigated opening of the burst membrane and subsequent thermal treatment of the battery cells show no satisfying results regarding solvent removal and process time. It is also

only applicable for cells with burst membrane, which requires additional pre-sorting. Therefore, safety measures have to be envisioned, if the solvents are separated either directly at or after cell opening. These options have to be applied in particular for pouch and round cells independent of their size and mass. For those cells can be expected that the solvent release during cell opening is lower compared to prismatic cells due to higher energy utilization.

Manual cell opening and dismantling to electrodes level simplifies further material separation, but is a time consuming, and thus an uneconomic process (Pinegar et al. 2020). Furthermore, it lacks to scale-up for the increasing variety of battery geometries and types (Wang et al. 2016). Therefore, dismantling to cell or even module level only are more realistic scenarios for high-throughputs in industrial applications.

In general, the safe cell opening depends on the batteries´ state-of-charge and health, the deactivation status of flammable organic solvents. If RE has to be maximized, plastics, electrolyte, graphite and Al have to be recovered additionally to Cu, Li metal oxides and steel. Thus, discharging is essential for cell opening and the solvents have to be extracted at temperatures at which the conductive salt and plastics do not decompose. The further separation of the solvents into their respective components after extraction from the batteries remains an unsolved task.

Several industrial applications are available worldwide providing secondary raw materials for battery applications or other products. One concept is the direct reuse of cathode active materials for new batteries (Shi et al. 2018). However, this reuse is not yet proven to be feasible with the continuous development of battery technology so far (Larouche et al. 2020). Alternatively, the design for recycling could be promoted enhancing the manual or even automatic dismantling and discharging of the batteries. However, cell opening and separation of the individual components remain the bottlenecks for automation from a manufacturing processing point of view (Marshall et al. 2020).

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Contribution

D.M.W. designed and carried out the experiments. D.M.W. prepared the manuscript and analysed the data integrating contributions from T.M. and U.A.P. T. M. and U.A.P. critically reviewed the work, the former multiple times.

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Conflicts of Interest:

The authors declare no conflict of interest.

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