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Systematic Identification of Hazardous States and Approach for Condition Monitoring in the Context of Li-ion Battery Disassembly

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Abstract

By increasingly strict regulations, the automotive industry is forced to create environmentally friendly vehicle concepts such as battery electric vehicles (BEVs). The automated dismantling of Li-ion batteries up to cell level provides the opportunity to remanufacture or reuse the individual cells or recover raw materials through recycling. However, there are risks in the dismantling procedures which jeopardize a trouble-free process. These risks include the leakage of toxic gases, the release of chemicals or the occurrence of thermal runaways, which may pose a fire or explosion hazard if disassembled incorrectly. The main aim of this paper is therefore to systematically identify requirements for Li-ion battery condition monitoring during disassembly for recycling based on the hazard potential during this process. The key is to identify critical paths that will certainly lead to a hazardous condition to be able to react with countermeasures in time. For this purpose, all hazard potentials must first be identified. Secondly, their cause-effect principles can be modelled to demonstrate a broad overview of all hazardous path sequences and paths which may occur during the disassembling process. As a result, especially safety-critical path sequences and paths are worked out which are relevant for disassembly and subsequent recycling. Based on this step, possible measurement parameters are identified and a proposal for a condition monitoring concept is made.

Keywords: Disassembly; Manufacturing system; Li-ion battery monitoring

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1. Introduction

The automotive industry is driven by increasingly strict regulations and the desire to establish environmentally friendly mobility concepts such as electric vehicles. Many automotive OEMs accelerated their endeavours and constantly released new battery electric vehicles (BEVs) using traction batteries [1]. However, due to a continuous electrification of vehicles, the ever-rising awareness regarding a sustainable utilization of valuable and scarce raw materials and end of life (EOL) scenarios including remanufacturing, recycling and reuse has intensified [2, 3, 4, 5]. Consequently, the establishment of closed-loop models in the context of battery production is crucial. Hereby, the automated dismantling of Li-ion batteries up to cell level provides a unique opportunity of recovering most of the raw materials used and achieving one of three crucial proceedings: reusing, remanufacturing, and recycling [3, 4]. Overall, the economic and ecological necessity to increase the disassembly granularity down to cell level is inevitable to save scarce raw materials and functioning components, thus ensure a sustainable handling.

However, battery packs as well as battery modules show a particularly high diversity in types and specifications which substantially complicates the disassembly process ([5] or [6]). More importantly, existing research illustrates solvable and unsolvable joining connections [6]. This is of high relevance for the disassembly process as unsolvable joining connections (e.g., riveted, welded) require destructive disassembly steps (e.g., cutting) which can lead to hazardous conditions [6, 7, 8]. For instance, the separation of non-detachable joints through cutting processes may have critical influence factors such as heat or forces [7, 8]. Those risks jeopardise a trouble-free dismantling process and can be clustered into thermal, chemical, electrical, and mechanical nature. They include the generation

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of toxic gases, the release of chemicals or the danger of a so-called thermal runaway, which poses a fire and explosion hazard if disassembled incorrectly. Existing research in the area of hazardous disassembling processes of Li-ion battery modules is either looking into specific dangers such as the thermal runaway [9] or into individual or several of these hazardous areas (e.g., chemical and electrical [9], thermal, electrical and mechanical [10]). However, there is a scarcity of research giving a broad picture of all existing hazardous areas by identifying and analysing the impact of the core processes of the disassembly on those hazardous conditions. The main aim of this paper is to systematically identify requirements for Li-ion battery condition monitoring during disassembly for recycling based on the hazard potential during dismantling of Li-ion batteries. For this reason, all hazard potentials and conditions must first be identified, recorded, and their cause-effect principles must be modelled to fully understand the entire safety related disassembly process hazards. In a subsequent step, safety critical paths are identified and potential measurement parameters are derived. Lastly, a proposal for a monitoring concept is made.

2. Fundamentals of Li-ion batteries

Li-ion batteries are present in various areas and are increasingly used as traction batteries in the automotive industry due to their advantages such as high power and energy density [11, 12]. A common topology concept has been proven in recent years, consisting of three hierarchical levels: battery pack, module, and cell. As a rule, individual battery cells are interconnected to form a battery module, which in turn is installed in a battery pack as highest level. [12] The medium level, and subject of the concepts of this paper, is the battery module which essentially consists of the following components: cell, cell contacting, thermal management, module housing, cell fixation and battery management system (BMS) [13].

Battery cells as the core of any battery system store electrical energy that will ultimately provide the energy to the e-motor to power the vehicle. Generally, Li-ion battery cells are built up of a composite of anode, separator, cathode, an electrolyte, current collectors, and a housing. [14] Anode and cathode usually consist of active material which is coated on a thin metal foil. A common cell configuration is an anode consisting of a copper foil with coated graphite as active material, while the cathode comprises an aluminum foil coated with NMC ($LiNi_xCo_yMn_zO_2$) [11]. However, there are also several other cathode types with the most popular ones being LCO ($LiCoO_2$), LFP ($LiFePO_4$), LMO ($LiMn_2O_4$), NCA ($Li(Ni,Co,Al)O_2$) [5]. All of them have in common that they present a risk if being exposed to critical conditions (e.g., temperature rise). Moreover, the permeable separator separates the anode from the cathode and the electrolyte is usually made of a non-aqueous solvent (e.g., $C_3H_4O_3$) and solution salt LiPF6 [15]. In general, cells can be classified into different types: cylindrical, prismatic and pouch cells [11]. Cylindrical and prismatic battery cells consist of a hard case (e.g., aluminum, stainless steel). An electrode jelly roll is inserted into those hard cases and is then welded [15]. In the case of pouch cells, the electrode stack is often produced by z-folding or stacking. Pouch cells are enclosed by a form-instable pouch foil which is sealed in a heat-sealing process [11, 15]. However, product and process related dangers can be identified and mapped independently from the cell type as examined in the following chapter.

3. Identification of Product Danger and Cause-Effect Chain

This research paper uses the methodological approach of concept mapping following the idea of Novak [16]. A concept map is a representation of a complex situation and shows explicitly all the relations between concepts, respectively being called nodes, using linking propositions between each concept and arranging those ideas in a hierarchical form [16]. Furthermore, complex situations are broken down into their key elements to illustrate the big picture of all hazardous situations concerning Li-ion batteries during the disassembly process. The causal chain concept map derived in this research paper, as illustrated in Fig. 1, consists of two axial interceptions comparable to a table. The first section on the left side illustrates the hazardous divisions comprising of thermal, chemical, electrical, and mechanical. These four categories are collaboratively described as crucial battery cell and module related characteristics by various sources (cf. Table 1). However, the following Table 1 shows that state of the art scientific research has not yet deeply analysed all four categories regarding internal effects and external consequences within one work. This paper merges all mentioned hazardous areas of battery modules and therewith allows to investigate all major critical states and paths which may possibly occur during the disassembly process.

Table 1. State of the Art of scientific research of battery cell module and pack related characteristics by various sources

Sources	**therm.**	**chem.**	**electr.**	**mech.**
Korthauer (2013)		X	X	X
Thiel et al. (2018)	X			X
Lisbona & Snee (2011)	X		X	X
Rothermehl & Nowak (2016)	X	X		
Diekmann et al. (2018)		X	X	

The individual columns on the top of Fig.1 illustrate the cause-effect chain comprising of an external influence factor (cause) followed by internal effects and external as well as human and environmental consequences. During the disassembly process several external factors, such as for example an external heat, supply may jeopardize the safe state of a battery module and the battery cell. Consequently, those causes can affect the battery cell and entail major issues up to a fire or explosion in the worst case [17, 18, 19]. Hereby, a battery module without production defects is assumed since initial production defects of the cells or module are expected to cause problems even before the disassembly. In the following, each path sequence starting with external factors will be described and explained in more

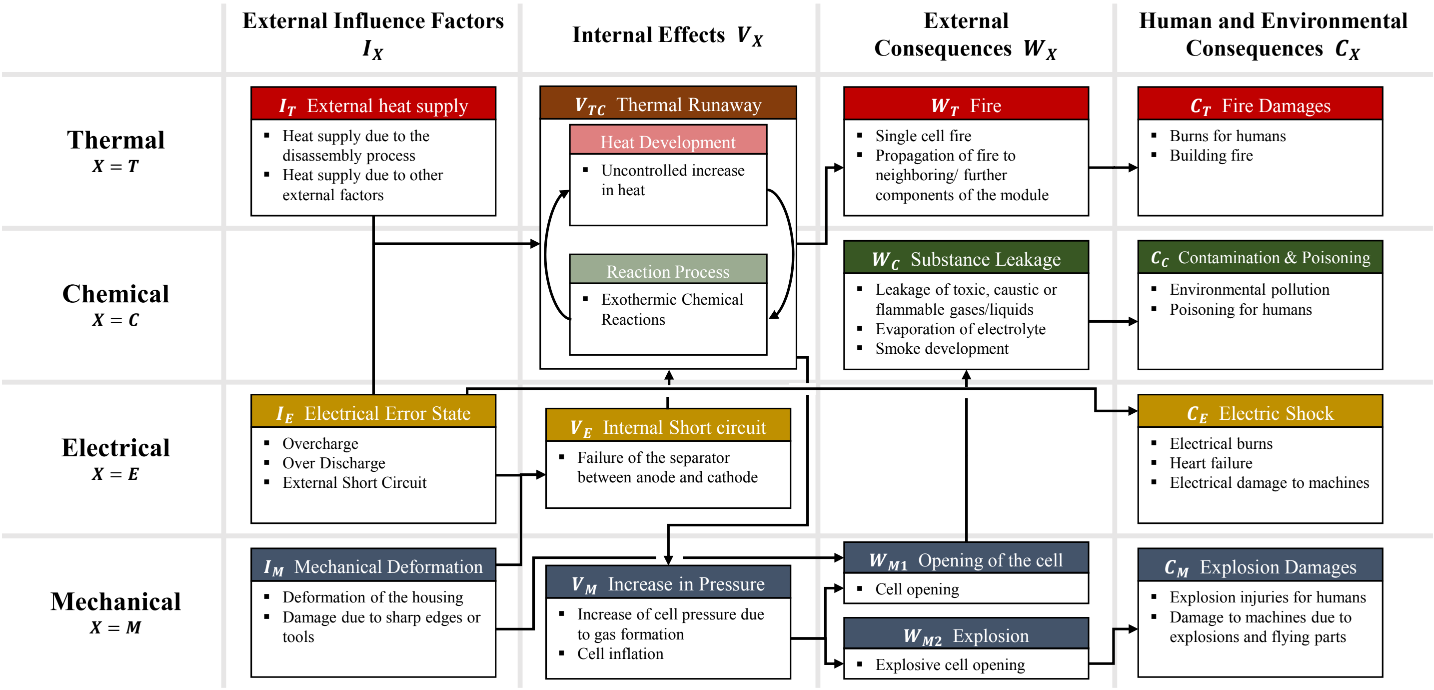

Fig. 1. Cause-effect chain of Li-ion battery hazards

detail. It is important to note that successive path sequences of different events from the areas of ”external influencing factors”, ”internal effects”, ”external consequences” and ”consequences for people and the environment” are referred to as paths (e.g., external heat supply leads to thermal runaway leads to fire and fire damage). One path sequence is defined as a link of only two events, e.g., electrical error state leads to internal short circuit.

3.1. Thermal process

Thermal processes usually start with an external heat supply as external causing factor. Critical external heat supply might result from the disassembly process itself or from other external factors (e.g., temperature in the production site, air temperature, induced heat through the use of tools or failure of cooling) [17]. The consequences of external heat supply are large independent of its cause: The increase in cell temperature is followed by a thermal decomposition of further components which again causes additional heat development [14, 20]. This described phenomenon of a self-accelerating decomposition process illustrates the mentioned thermal runaway. Precisely, a thermal runaway comprises an almost indefinite process of heat development due to uncontrolled increase in temperature (thermal process) and a chemical reaction process due to decomposition processes (chemical process) [14]. The consequences of a thermal runaway are very often uncontrolled cell heating up to cell fire [21]. A spread of fire to neighboring components of the battery module could possibly follow. Ultimately, such a cell fire causes several problems for the human or the environment: The risk of burns to employees and humans arises and fire or damages to buildings or machines can occur [9, 19]. Besides the uncontrolled cell heating, the second possible consequence of a thermal runaway is a pressure rise due to an increasing gas formation within the battery cell and a potential inflating of the cell [14]. Again, this behavior can directly cause an opening of the cell which in turn causes a material leakage followed by a potential contamination or poisoning or secondly an explosion and damages (cf. subchapters 3.3 and 3.4).

3.2. Chemical processes

The second section comprises all chemical processes. Generally, there has not been identified any case in which a chemical process presents an external factor. However, one step further in the internal effect category, chemical processes are a major part of the thermal runaway as described in subchapter 3.1. The chemical safety of the Li-ion battery primarily depends on the thermal stability of the cell components and their degradation potential. Typically, decomposition substances can consist of flammable gases such as hydrogen (H_2), carbon monoxide (CO), methane (CH_4), ethane (C_2H_6), toxic compounds such as CO, hydrogen fluoride (HF), monophosphane (PH_3), as well as hazardous and harmful substances such as aldehyde and carcinogenic dusts from cathode oxides of cobalt and nickel. At the same time many cathode materials release oxygens. The electrolyte is the main source of gas evolution, resulting from reduction at the anode or oxidation at the cathode. [10, 14, 22]

The paths directly following a thermal runaway are entirely non-chemical processes. However, at a later stage after mechanical effects (increase in pressure and cell opening), a thermal runaway may also cause indirectly another chemical process, namely the leakage of substances [14]. The leakage can comprise of toxic and highly flammable gases, electrolytes, or other toxic and dangerous substrates [10]. Once the external consequence of a substance leakage is reached, the only following step is the contamination and/or poisoning, which poses an enormous danger for both the human and the environment [23]. Lastly, another way to get to a substance leakage is through an entirely mechanical process of mechanical deformation followed by an increase in pressure until the cell opens and substances may leak out (c.f. subchapter 3.4).

3.3. Electrical processes

The third sector, namely the electrical section, is one of the most causing and therewith dangerous external factors as it can entail the entire effects and consequences of the causal chain concept map via direct and/or indirect paths. The starting point of the electrical section is an electrical error status such as for example overcharging, over discharge or external short circuits [14, 24, 25]. Those external influential factors can in turn have three different consequences. Firstly, it may cause a thermal runaway as described in detail in subchapters 3.1 and 3.2. Secondly, an electrical error status can have an internal short circuit as consequence, which means that the separator between the anode and cathode fails [18, 25]. The only subsequent state will be again a thermal runaway [25, 26]. Thirdly, an electrical error status may imply an electrical shock as a very dangerous consequence for human beings [21].

3.4. Mechanical processes

The fourth and last hazardous division is of mechanical nature. Deformation of the battery module or cell housing presents a mechanical deformation and therewith an external influence factor. The consequences can be twofold: Firstly, an internal short circuit may occur as an internal effect [26]. Secondly, as an external result, one or several battery cells of a battery module might open [26]. The former case, internal short circuit, is described in the previous sections in detail as it always entails a thermal runaway. The latter case, opening of the cell, results in a potential substance leakage. This external consequence could lead to a contamination or poisoning (c.f. subchapter 3.2). However, besides those paths being directly linked to a mechanical deformation as an external influence factor, there is another internal consequence, namely the increase in pressure, which is being triggered by the path coming from a thermal runaway (cf. subchapter 3.2). This means that a thermal runaway can cause an increase of pressure within the cell due to gas formations. Potentially, the cell even inflates due to a high rise in pressure [14]. This internal effect can have two external consequences: Either an opening of the cell as described above or an explosion (e.g., explosive opening of the cell). The latter case may cause immediate damages to machines or damages due to flying parts as well as possible injuries to humans.

4. Identification of Critical Paths and relevant Hazardous States for Battery Disassembly

The present chapter examines paths leading to a critical event. Critical events are illustrated in column 4 of Fig.1. However, by means of the cause-effect chain each critical event is triggered by an initial external influence factor (column 1 of Fig.1). Hazardous states refer to states that are subsequent to initial external influence factors, lie on one or more critical paths and, if no countermeasures are taken, lead to such critical events (consequences for humans and the environment). Consequently, hazardous states can be found in columns 2 and 3 of Fig.1.

For process assurance, it is necessary to derive all possible critical paths when dealing with batteries and to determine all critical paths relevant in battery disassembly for recycling purposes. Based on Fig.1, the set of all possible critical paths can be derived. We denote I_x as influence factors of the causal chain, where $X \in [T, E, M]$ and where T denotes the thermal influence, E the electrical influence and M the mechanical influence. Critical events are denoted with C_X with $X \in [T, C, E, M]$ with T being fire damage, M being explosion damage, E being electrical shock and C representing contamination. Furthermore, V_{TC} denotes the thermal runaway. All derived critical paths are derived and summed up in mathematical notations (see Table 2).

Based on the set of all critical paths, those relevant for battery disassembly for recycling are extracted and marked with "X" in Table 2. Before Li-ion batteries are disassembled for recycling purposes, discharging of the Li-ion cells for example through salt solutions or by direct Ohmic discharge takes place [5]. Hence, critical paths starting from I_E or containing V_E can be ruled out.

Table 2. Critical paths of cause-effect chain of Li-ion battery hazards

Path No.	**Critical Path**	
1.	$I_T \mid I_E \rightarrow V_{TC} \rightarrow W_T \rightarrow C_T$	X
2.	$I_T \mid I_E \rightarrow V_{TC} \rightarrow V_M \rightarrow W_{M2} \rightarrow C_M$	X
3.	$I_T \mid I_E \rightarrow V_{TC} \rightarrow V_M \rightarrow W_{M1} \rightarrow W_C \rightarrow C_c$	X
4.	$I_E \rightarrow V_E \rightarrow V_{TC} \rightarrow W_T \rightarrow C_T$	
5.	$I_E \rightarrow V_E \rightarrow V_{TC} \rightarrow V_M \rightarrow W_{M2} \rightarrow C_M$	
6.	$I_E \rightarrow V_E \rightarrow V_{TC} \rightarrow V_M \rightarrow W_{M1} \rightarrow W_C \rightarrow C_c$	
7.	$I_E \rightarrow C_S$	
8.	$I_M \rightarrow W_{M1} \rightarrow W_C \rightarrow C_c$	X
9.	$I_M \rightarrow V_E \rightarrow V_{TC} \rightarrow W_T \rightarrow C_T$	
10.	$I_M \rightarrow V_E \rightarrow V_{TC} \rightarrow V_M \rightarrow W_{M1} \rightarrow W_C \rightarrow C_c$	

The resulting paths can be deemed relevant for battery disassembly for recycling purposes. During the actual process of disassembly, mainly handling and separation operations take place [7]. Both handling operations and, in particular, mechanical separation operations (e.g., milling open of non-detachable joints) can result in mechanical deformation. Accordingly, the critical path starting from I_M, which ultimately entails contamination of the environment C_C due to escaping substances, must be considered. In addition to mechanical deformation, separation processes also cause an external heat input [8]. If a critical heat input is exceeded, this can lead to numerous critical paths, starting from the initial external influence I_T. In summary, the relevant critical paths for Li-ion battery disassembly for recycling that must be prevented from reaching their respective critical state are path no. 1, 2, 3 and 8 which are marked with ”X” in Table 2.

From the derived relevant critical paths, it can be deduced that there are also hazards for the environment after a deep discharge. Except for the electric shock, all critical events can still occur. 3 of 4 critical paths (critical paths no. 1-3), which lead to fire damage, explosion damage and contamination damage, pass through the thermal runaway as an intermediate state. Furthermore, there is a critical path that leads to a critical end state without the thermal runaway (critical path no. 8). Therefore, thermal runaway and the opening of the cell, which immediately leads to the leakage of substances, are identified as critical states within the critical pathways whose occurrence must be monitored by a condition monitoring system.

5. Proposed Condition Monitoring Concept for the Disassembly of Battery Modules

Existing methods for condition monitoring of thermal runaway for Li-ion battery modules can be roughly separated by the location of the sensor for measuring the variables that provide information about the occurrence of a thermal runaway [34, 36]. On the one hand, external measurements are possible, in which sensors are attached outside of the battery module. On the other hand, sensors installed in the module are also mentioned. These include internal temperature sensors, voltage and current sensors, resistance sensors and pressure sensors. Especially for deeply discharged cells, as they are considered during disassembly, the approaches that work with internal sensors can be excluded, as no power supply or the interruption of the current and information circuits is the consequence.

For a monitoring concept, it is therefore necessary to work with external sensors, especially if battery modules from different manufacturers are to be dismantled, since there is no immediate access to the battery management system. Additionally, since a thermal runaway is characterised by a specific heating process of the respective cell or the affected cell groups, targeted monitoring of the heating state of the entire battery module is a possible approach to monitor relevant critical paths 1-3.

The heating process in a thermal runaway is largely dependent on several factors. These include the specific properties of the cell (material, geometry, ...), safety mechanisms installed in the module and the history of the cell or battery module. Therefore, the critical temperature that triggers a thermal runaway of the discharged Li-ion battery cells must be determined individually for each cell type which are built in the battery module. This can be carried out through experimental tests for example using Accelerating Rate Calorimeter.

The monitoring concept proposed in this work is depicted in Fig. 2. An externally mounted thermography camera and a weight measuring system are used to detect the thermal runaway and the substance leakage. On the one hand, the thermography camera enables the monitoring of a maximum permissible limit temperature of the battery module. Furthermore, it is also possible to evaluate the absolute or relative temperature curve over time, which allows conclusions to be drawn about critical heating processes of the module. In this context the correlation between the actual internal heating of individual cells affected by thermal runaway and the time-delayed detection on cell or module surfaces is of special importance. Regions of interest must be defined as cell or module surfaces where the occurrence of a thermal runaway due to heating of the cells becomes visible without delay. For this purpose, temperature-controllable dummy cells will be developed in subsequent work to simulate the thermal runaway. This will enable investigations into the thermal detectability of the thermal runaway.

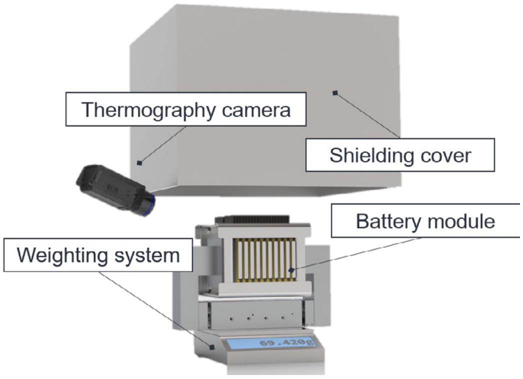

Fig. 2. Proposed condition monitoring concept consisting of a thermography camera, weighting system and shielding cover

The weight detection system pursues a similar goal as the thermography camera. In addition to cell heating, a creeping decrease in weight is an indication of escaping substances and thus an opened cell. Also, the weight gradient can be recorded.

If a critical state is detected, appropriate remedial measures must be taken. In the concept developed in this work, a shielding cover is shown above the battery module in Fig. 2. It is lowered onto the battery module when e.g. a thermal runaway is detected, so that the module is shielded from the environment and can burn out in a controlled manner.

6. Conclusion and Outlook

Within this paper, requirements for a condition monitoring system for Li-ion battery disassembly in the context of recycling are derived by systematically analyzing the hazard potentials and modelling the cause-effect chain. Firstly, four major hazard potential divisions (thermal, chemical, electrical, and mechanical) are identified and their cause-effect chain comprising of external influence factor (cause) followed by internal and external effects as well as human and environmental consequences are further determined. The entire process is modelled using the methodological approach of a concept mapping giving the possibility to identify critical paths. In a following step, relevant critical paths of Li-ion disassembly for recycling are derived. Using early-stage detection could prevent those critical paths from occurring. Finally, a condition monitoring concept for detecting thermal runaway and substance leakage by using a thermography camera and weighting system is given.

In further works, experiments to characterize the thermal runaway of variants of different battery cells will be performed. Furthermore, testing and calibration of the proposed condition monitoring concept using a heatable dummy battery cell as well as the weight measuring system will be carried out. The validated monitoring system will then be integrated into an existing Li-ion battery disassembly system.

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