Chapter 1. Lifecycle and sustainability

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Résumé

Les batteries de type lithium connaissent et vont connaitre un essor considérable compte tenu d'une part de leurs bonnes performances et d'autre part d'une demande sans cesse croissante d'énergie notamment pour les applications transports. Cet accroissement entraîne une consommation accrue de matières premières et exige, dès aujourd'hui, de penser « cycle de vie » et « développement durable » afin de préserver et de pérenniser les ressources naturelles. Ce chapitre décrit dans un premier temps la méthodologie de l'« analyse du cycle de vie » appliquée aux batteries au lithium à partir de l'analyse de travaux publiés dans la littérature. En particulier les points clefs de ces études sont soulignés et des améliorations indispensables dans l'application de la méthode sont proposées. Dans un deuxième temps, le chapitre décrit un rapide état de l'art du recyclage et pointe la nécessité d'intégrer et de modéliser tout le cycle de vie des batteries depuis l'extraction des ressource primaires jusqu'à la fin de vie.

Abstract

The actual and future rapid development of lithium batteries is caused by both their good performances and by increasing energy demand, especially for transport applications. This implies a growing consumption of raw materials. Taking into account this rapid growth, it is necessary, from now on, to think “life cycle” and sustainability in order to preserve and to sustain our natural resources. Based on a literature review, this chapter presents at first the life cycle assessment (LCA) methodology as applied to lithium batteries. Especially the key points of the analyzed studies will be emphasized here and some improvements will be proposed. The end-of-life stage is analyzed then and a short summary of existing recycling methods is given. Finally it will be emphasized that the environmental impact assessment of future lithium batteries should be done by integrating the entire battery life cycle from ressources extraction up to recycling.

Key words

Batterie lithium, Analyse du cycle de vie, durabilité, recyclage

Lithium Batteries, Life cycle analysis, sustainability, recycling

Introduction

The concept of sustainability was developed in order to improve the present human living standards while maintaining the availability of the natural resources for the future generations. According to this definition, technological development is a way to improve the sustainability, because it enables to meet human needs by transforming natural resources into useful products [1] .

By 2050, the urban world population is expected to approximately double to an estimated 6.4 billion [2] and we are aware that the Earth natural resources are already limited. In this context, less impacting and more efficient industrial processes' design represents a real challenge for engineers. From now on, the impacts of new technologies have to be assessed in detail, all along their life cycle, even before their massive industrial deployment. We should be sure that the generated impacts are actually counterbalanced by the improvement of the living standards on Earth.

In this chapter, we will consider new technologies related to the development and treatment of lithium batteries. In the first part, we will demonstrate how existing studies are already taking into account environmental impacts assessment and we will particularly emphasize the main assumptions realized using life cycle assessment (LCA) approaches. In the second part, we will focus on the end of life of lithium batteries to demonstrate that the entire value chain has to be considered while arbitrating on the acceptability or not of a design decision from an environmental perspective.

1. Life cycle assessment applied to lithium batteries "Concept, method and key-results".

According to the United States Geological Survey (USGS) lithium battery market is expected to increase by approximately 200% by 2017 [3] and the main application of this technology would be electric/hybrid vehicles. This incoming technology is apparently environmentally friendly because of its zero-emissions during utilization phase due to the absence of any combustion processes. Nevertheless a closer look is needed in order to understand the impacts of the battery throughout the entire product life cycle, from minerals' extraction step to its end-of-life, and not only during the use phase. Thus, the question raised here is, whether the use of lithium batteries in electric cars will provide a real environmental benefit compared to the former solutions.

To answer this question, a life-cycle assessment (LCA) has to be realised, taking into account all the steps (or stages) of the product life-cycle, by determining the amounts of energy consumed, mass balance of the all the components and by quantifying all emissions and wastes generated by the battery all along its life span. The use of the LCA methodology gives a multi-criteria vision of the different environmental impacts generated by the products or services considered (e.g. ozone depletion, global warming, raw material consumption, etc.). This approach facilitates also the comparison between them. Actually, it can be used during the design process to make decision and improve the products, services or organisations under design from an environmental point of view.

LCA is a standardized methodology described in the ISO 14000 environmental management standards. According to the ISO 14040 series an LCA is carried out in four iterative steps: **goal and scope**, **life cycle inventory**, **life cycle impact assessment**, and **interpretation** (Figure 1).

Insert Figure1

In the following paragraph, the objectives of each phase of the life cycle analysis will be explained and illustrated with different lithium batteries LCAs published in the literature. Then the key points issued from those LCAs will be raised here in order to improve the future lithium batteries LCAs.

To illustrate the four steps of lithium battery LCA six major scientific contributions [5-10] will be analysed in the following sections. The Table 1 synthetises the main characteristics of the six selected studies, namely:

- The functional unit that has been chosen for each case,
- The cathode chemistry of the solution under assessment,
- The life cycle phases considered,
- Key characteristics for each product.

Insert table 1

These studies are difficult to compare as their focus was not the same and as various assumptions were made by the different authors. Owing to this conclusion an analysis will be provided here for each step of the LCA methodology in order to guide future LCA studies.

1.1 Goal and scope definition

The goal and scope definition step aims to define following items: the functional unit which is used as a basis for the comparison of several solutions; system boundaries; impact categories chosen; allocation methods used to partition the environmental load of a process when several products or functions share the same process. These elements will be further discussed in the following paragraphs.

Functional unit definition

The functional unit (FU) is based on the service provided by the product/system/solution and focuses on the main functionalities. It allows the comparison between several solutions that provide the targeted service. The good practices for functional unit definition specify that a well-defined functional unit shall contain an infinitive of a verb to define the service provided, a technical criteria that qualifies the performance of the system and an operating time for the whole life-cycle.

[5, 6, 7, 9] studies have defined functional unit that is conform to the best practices stated above. Majeau-Bettez et al. [6] and Zackrisson et al. [7] define their functional unit in terms of quantity of energy stored in the battery (and provided to the vehicle) during the life time of a vehicle. Majeau-Bettez and al. [6] directly define the functional unit in terms of energy stored and release during a charge and discharge cycle. Zackrisson et al. [7] use a definition which is related to the batteries performance, whereas the capacity of the battery, the depth of discharge and the number of charge discharge cycles are given. EPA researchers [5] and Notter et al. [9] define their functional unit as a driven distance over the average life span of a car vehicle (around 200 000km). “km driven” based functional unit present the main advantage to allow an immediate comparison of the results with other systems that provide energy to vehicles (like petrol or hybrid systems or fuel cells). In the other hands, the use of this kind of functional unit will not permit the comparison with batteries used for very different applications like those used for off grid systems. For the purpose of comparing batteries used in various applications, “energy stored” based functional units enable a direct comparison.

Unfortunately there is no consensus about the choice of a FU, even among the researchers working in the same science field. Consequently, it is recommended to provide all the performance criteria that allow converting the selected functional unit into another.

System boundaries identification

Providing the service stated by the functional unit generally requires a lot of activities along the product life cycle. The aim of system boundaries definition is to state which activities are actually taken into account in the study and which are insignificant or constant for the solutions compared. Actually, excluding a significant activity from the inside of the system boundaries might hide some transfer of environmental impacts. Thereby, a clear justification or explicit risks related to this decision have to be given in the final LCA report. These boundaries can be defined at two levels. At first, the decision is taken about the life cycles stages which will be included or not in the study. Next, for each life-cycle stage included in the study it is also necessary to specify which activities are included in the model.

Concerning the life-cycle stages, the analysis of the six publications reveals that six stages have to be considered for the life-cycle assessment of a lithium battery pack. These stages are:

- Extraction of raw materials required for the entire battery system (including BMS, passive cooling system etc.). These material includes Lithium, Aluminium, Copper and some others rare and heavy metals like Cobalt, Manganese, …
- Active material processing including all the production steps of anode, cathode, electrolyte and separator materials required to assemble a battery cell.
- Manufacturing of battery stage encloses all the required activities to assemble a battery cell from active materials and the processes used to construct a battery module from the cells.
- Manufacturing of additional components like cooling system, battery management system (BMS), packaging, …
- Use phase in the vehicle including the energy necessary to charge the battery.
- End of life strategies.

The table 2 synthetises the stages taken into account by the different authors.

Insert table 2

The details concerning the choice of the activities included in each life-cycle stages are not given in all of the six analysed studies. Data from existing databases were surely used and the impacts related to the building of remanufacturing plants and the impacts related to the infrastructure (production of machine tools, lighting end heating of factories…) were certainly neglected too. However, some details are given concerning the end of life stage. Dunn et al. [8] have proposed a comparison between three different strategies, namely pyrometallurgy, hydrometallurgy, and direct physical recycling. EPA researchers [5] have built a mean end-of-life scenario which includes landfilling, metal recovery and incineration. Finally, Notter et al. [9] have considered the worst scenario to model the end of life of the battery (i.e. landfilling).

From the theoretical point of view, the wider the boundaries are the less chances there are to hide an impact transfer. Consequently, in order to avoid impact transfer, the LCA practitioner would like to choose wide boundaries. On the other hand the amount of the work required to collect data and the associated uncertainties grow rapidly when boundaries of the system go wider. As a consequence, the boundary definition must be relevant to the studies expectations (comparison with other studies, design activities, etc.).

In the particular case of lithium batteries it seems irrelevant to exclude any life-cycle stage of the study area. As a matter of fact, the six life-cycle stages proposed previously for the assessment of lithium batteries environmental impacts would generate significant impacts. Excluding one of them from the scope of the study would highly raise the probability to hide an eventual significant impact transfer. More specifically, the end of life stage has been excluded from the scope of three studies [6][7][10] and the three others [5][8][9] have used models that are not comparable. As the end-of-life of lithium batteries seems to be a strategic issue a deeper model of the different end-of-life possibilities (i.e. open and close loop recycling, remanufacturing, incineration, landfilling) should be done.

Impact categories choice

According to the purpose of the study and system under assessment, different environmental impacts can be retained and different methods exist to estimate these impacts. The table 3 shows the environmental indicators used in the reviewed studies. Global Warming Potential (GWP) and Energy consumption (Energy) as indicators are used in all studies. Moreover, in the majority of the studies, Abiotic depletion potential (ADP), Acidification, Eutrophication and some Ecological Toxicity indicators are also used.

Insert table 3

Amongst all existing impact categories, some of them are more pertinent to illustrate the environmental impact of a lithium battery. The most appropriate indicators are discuss below:

- Batteries are devices made to store energy. Consequently it seems relevant to assess the ***energy*** needed along the life-cycle in order to assess the performance of the system regarding the energy the battery is able to release during its use phase.
- Thought that batteries are supposed to replace a combustion technology, ***Global Warming Potential*** is a good indicator to compare these two technologies. It also provides an idea of fossil-energy dependence of the system during its life-cycle.
- As batteries often contains some strategic metals like cobalt, lithium, copper and others, ***Abiotic Depletion Potential*** is a good indicator to quantify the scarcity of these resources as an environmental impacts.
- Most of the industrial processes for manufacturing and treatments at the end of life uses acid attacks/leaching. Consequently, assessing ***acidification*** seems relevant.
- In order to take into account impacts to the ecosystem of phosphoric substances like LFP, ***Eutrophication Potential*** is used.

- The chemical processes use various toxic substances; consequently assessing eco-toxicity indicators is relevant (***Marine Ecotoxicity Potential, Freshwater Ecotoxicity Potential, Human Toxicity Potential***).

Depending on the objective of the assessment, the most relevant indicators, as those already used in the previous studies, can be retained. It is generally recommended not to exceed more than 5 to 6 indicators in order to facilitate the results interpretation. Nevertheless, it is also necessary to check quickly the other indicators, just to control if there is no impact transfer between indicators when redesigning the model.

Allocation

In the industry, some production equipements might be shared between several products. For instance, the dry room required for the assembly of battery cells generally contains several production chains of different technologies. In that case LCA practitioners need to assign a part of the inventory of the shared process to its inventory. This operation could be critical if the shared process has significant impacts.

Moreover, another difficulty faced by researchers is the scale of the experiment: the same process running at the laboratory scale is generally more energy and material consuming than at industrial scale as some products could be re-used several times in closed loops; some reactors might be continuous, etc. Some key rules have then to be established and shared by the allocation and today no information is available on this aspect in the different studies.

1.2 Inventory Analysis

Inventory analysis consists in explicitly list all the activities that are inside the system boundaries and which are required for providing the service described by the functional unit.

LCA practitioners are invited to establish the list of processes included in all the product life cycle stages. Then as schematized in figure 2, for each process, it is necessary to quantify its:

- inputs: raw material and energy necessary,
- outputs: emissions and products resulting from the process.

Insert Figure2

Greatest part of the LCA practitioner's work is to select relevant data to use and to create appropriate life-cycle scenarios with the processes. One can distinguish two types of data used in an LCA: primary data and secondary data. Primary data states for data that are collected especially for the purpose of the study. These data have the advantage to be relevant to the study purpose because they are especially collected for. All other data are called secondary data. Secondary data can come from various sources such as bibliography, manufacturers, environmental databases, etc. Using secondary data is often time saving but needs controls to be sure that the data are relevant to the study purpose.

From all the six LCAs selected [5-10] all of them have used the secondary data. Researchers from EPA [5] are the only who have collected primary data. These data are those used for the active material processing and battery manufacturing which are, according to them, the cores of the process. All the other data of this study come from secondary sources.

Because of the recent interest for some rare resources on earth, the secondary data concerning these rare resources are expected to evolve in a near future and the indicators related to their scarcity would probably evolve too in order to better integrate their environmental impact.

1.3 Impact Assessment

Substances present on the inventory list cause generally certain environmental impacts that can be quantified and grouped into so-called environmental indicators, whereas each indicator is specifically related to a physical or biological phenomenon. For a given environmental indicator, each substance has a different contribution. For instance the global warming potential (*GWP*) can be calculated using the following equation :

$$GWP = \sum_i GWP_i \cdot m_i$$

where GWP_i is the individual Global warming potential of each substance released and m_i stands for the quantity of the i^{th} substance released in kg. Individual Global Warming potential of each substance can be found on the IPCC report[1]. For instance, figure 3 shows that the individual GWP of Methane (CH_4) is 25.

Moreover, a given substance can contribute to several indicators at the same time (figure 3). The calculation of the value of the indicators is always coded as a calculation method in LCA softwares.

Insert Figure3

Impact Indicators can be classified in two categories; midpoints and endpoints impacts as shown in Figure4. Midpoints indicators are grouped by the nature of the physical effect induced by the release (or consumption) of the substance. For example Global Warming Potential, Ozone Layer Depletion and Acidification Potential are Midpoint indicators. Endpoint indicators, indicators are grouped according to the effect they produce on their environment. For example Human Health, Ecosystem Quality and Resources are endpoint

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[1]IPCC Third Assessment Report "**Climate Change 2001**"

indicators. In a general, environmental indicators are not comparable among them because they are linked to very different physical effects (and uses different units).

Finally these three endpoint impact categories can be grouped in a single score. This is the last indicator that resumes all the previous indicators. It has the advantage of being easily comprehensible but it lacks accuracy, therefore the midpoints are generally more often used in the scientific community than endpoints and the single score indicator.

Insert Figure4

As mentioned previously, the choice of the indicators depends on the objective of the study. In Table 4, are summarised the choices realised by the researchers in the selected studies.

Insert table 4

Certain calculation methods include a limited number of midpoint indicators, so it may be necessary to use more than one method to include all the desired indicators. As an example CML calculation method includes the following midpoint indicators: Ozone layer depletion, Human toxicity, Fresh water aquatic ecotoxicity, Marine aquatic ecotoxicity, Terrestrial ecotoxicity, Photochemical oxidation, Global warming, Acidification, Abiotic depletion and Eutrophication [12].

Results of the impact assessment are usually represented by graphics, where the contribution of the given impact or the given life cycle stage to the indicator value is pointed out.

The figures 5 to 7 give an example of the representation of the results (extracted from Zackrisson et al.[7]) for all life cycle phases. The global warming potential was selected here as an example of an indicator (figure 5).

Insert Figure5

On the contrary in the Figure 6, life cycle impacts are presented for only one life cycle phase: the use phase, whereas the contribution of each cause (transport, weight, electricity losses) to the given impact is visualized.

Insert Figure6

Another classical way to present the LCA results is presented in Figure 7. The distribution of the global life cycle impacts as a function of the life cycle stages (transport, usage, and manufacturing) is shown here.

Insert Figure7

Thus, different focuses have to be done on the life cycle impact assessment results in order to emphasize the real causes of the impacts. Those figures have to be analyzed with respect to the initial objectives as defined in the first step of the LCA study in order to simplify the final interpretation.

1.4 Interpretation

The interpretation of a LCA study entirely depends on the 3 previous steps of the methodology (figure 1). From the six studies reviewed for this chapter, the following hotspots have been identified.

EPA [5] and Majeau-Bettez et al. [6] agree on the fact that Ni and Co extraction generates high impacts. This fact is confirmed by Notter et al. [9] and Ellingsen et al. [10] who also shows that metal extraction -like Cu and Al- induces high impacts.

EPA [5], Majeau-Bettez et al. [6] and Zackrisson et al. [7] agreed on the fact that electricity grid for use phase is an important parameter that raises several indicators, especially GWP since, in some regions of the earth, electricity power plants are using fossil fuel.

Dunn et al. [8], in agreement with the statement of Notter et al. [9], demonstrates the environmental benefit of lithium batteries' recycling. Unfortunately, recycling is not assessed in all the studies because the end-of-life (EOL) scenarios are not steady yet as there are several possible chemical compositions of lithium batteries. This point will be further discussed in the second part of the chapter.

Notter et al. [9], Majeau-Bettez et al. [6] and Ellingsen et al. [10] reported the importance of considering also the auxiliary elements of the battery pack inside the boundaries. For instance, they report that the Battery Management System that includes copper has significant environmental impacts. Ellingsen et al. [10] and the report of EPA [5] highlighted the importance of the electrode processing, especially of their active materials (for both the cathode and the anode). More in detail, Ellingsen et al. [10] and Zackrisson et al. [7] agree on the importance of the impacts generated by the NMP solvent used to process the cells' active material.

Finally it is also important to notice that Dunn et al. [8] and Ellingsen et al. [10] disagree on the quantitative assignation of the dry-room process. But as none of them explain in detail how this assignation is done, it is difficult for the reader to make his own opinion. This shows the importance to clearly state the hypotheses which are taken to make such assignations.

As the six selected studies do not share the same hypotheses concerning functional unit, boundaries and objectives, it is impossible to compare the obtained results. Moreover some of the studies have used the LCA to assess the performance of the batteries along the entire life cycle, whereas some of the studies have excluded certain stages (steps). In some cases, environmental benefits were emphasized with the use of several end-of life scenarios.

1.5 Conclusions

The manufacturing technology seems now to be mature and the batteries life cycle can be well described, even if certain processes are still evolving. Different studies have already assessed the environmental impact of lithium batteries' packs and this section has demonstrated that the way the LCA methodology is applied influences significantly the final results. But it is also highlighted that LCA can provide new indicators to guide the design of the Lithium batteries if the LCA is well conducted, that means if:

- The Functional Unit is well defined
- The impact indicators are carefully selected in relation of direct environmental concerns while some other indicators are just chosen to avoid impact transfers.
- The data collection is adapted with a clear allocation when needed
- The interpretation is clear and assumptions well defined.

The question now is: how to address the end of life of those products and particularly how to recover rare resources inside the batteries? Indeed, all the realized studies were developed on an existing or well defined solution with limited boundaries and without considering closed loop life cycle strategies. It seems thus necessary to enlarge the perimeter of the studies and to propose a model of both the product itself and the necessary industrial chains required to manufacture and recycle it. The objective here is to be able to choose the best compromise between the performances of the batteries, the manufacturing strategies and the recycling strategies. Such a model is required to avoid any impact transfer from an environmental impact to another impact or from one life cycle stage to another.

In the next section, the existing end of life processes for batteries will be presented. This section will also be identified parameters to be taken into account in the future lithium battery LCA model integrating the end of life processes.

2. From recycling process definition to sustainable industrial solutions

As discussed in the previous section, the recent emergence of hybrid and electric vehicles together with a rapid development of portable electronics has provoked increasing need for performant energy storage systems. Accordingly the world battery market is expanding. Researchers and industrials have thus focused their effort on the development of more performant rechargeable batteries. In particular, lithium-ion technology appears currently as the most performing system due to its energy density, cyclability and energy yield and tends to supplant older technologies, such as lead acid batteries, nickel metal hydride (Ni-MH) or nickel-cadmium (Ni-Cd), which should be banned soon. In European Union, the recycling of all used batteries is regulated by the 2006/66/EC directive [14]. This directive enforces the principle of extended producer responsibility and imposes recycling rates depending on the technology (see Table 5). In particular, according to the batteries technology, recycling rates of 65, 75 or 50 % by average weight of batteries and accumulators shall be achieved respectively for lead-acid, Ni-Cd and all other types of batteries. Furthermore it is recommended to attain the highest technically feasible degree of material recovery for cadmium and lead.

Insert table 5

Taking into account that the sources of raw materials are limited and that their supply could become uncertain or difficult due to geo-political tensions, the necessity is to ensure the sustainability of their supply for not only environmental but also economic reasons. This implies the establishment of an efficient recycling chain.

2.1 Recycling process

Lithium batteries' recycling process is nowadays essentially driven by economical profits [15], since the recovery of battery constituents (such as cobalt, nickel and copper) is cost-effective. Nevertheless good candidates for electric vehicles applications are cathode materials based on lithiated manganese or iron phosphate salts, which are less toxic and less expansive. This should be taken into account when designing their future recycling chain. This process should be whether less expensive than the actual processes or whole components (e.g. cathodes, anodes, electrolytes…) should be recovered instead of chemicals products as done currently in order to keep the so-called "added value".

The efficiency of the whole recycling chain is obviously depending on the efficiency of the recycling process itself but is also conditioned by the efficiency of both collection and sorting steps. Currently only metals are recovered from spent batteries and both pyro-metallurgical and hydro-metallurgical processes are employed. A combination of the two processes can also be used. Indeed, the pyro-metallurgical recycling process consists in a high temperature treatment, reaching around 1400°C, where battery cells are smelted in a furnace and afterwards valuable metals as cobalt, copper and nickel are recovered. Generally, neither aluminum nor lithium are recovered since it is not energetically/economically efficient and they leave the process in the form of a slag that can be used as an aggregate in concrete [16]; In such a process there is no need to sort batteries by chemical composition. The only necessary pretreatment is their dismantling and grinding. The pyro-metallurgical process is commercially exploited by UMICORE, in Belgium. Regarding the hydro-metallurgical recycling, the process is mainly used to recover the metals from battery's active materials (both cathode and anode) [16] and it is based on a leaching process, whereas different acids or bases or solvents can be used as leaching agents combined eventually with a precipitation/filtration step. Since it is a low-temperature process it is less energy consuming than pyro-metallurgical treatment. It enables the recovery of copper and other valuable metals as pyro-metallurgical process does, but aluminum and lithium salts can also be recovered and re-used later in the active material construction chain. On the other hand, this process would benefit from a higher degree of batteries' sorting prior to the recycling process. As well as for the pyro-metallurgical treatment, the batteries are dismantled at the beginning of the process. Some of current recycling processes combine both pyro-metallurgical and hydrometallurgical steps to recover more valuable metals. Each technique presents several disadvantages (energy consumption, waste water production …). A comparison of pros and cons of both processes is summarized in the figure 8.

Insert Figure8

As an alternative to both mentioned processes, the Direct Physical Recycling technique was proposed in the literature. This treatment deals with the recovery of battery constituents in order to be reinserted directly into the battery supply chain with little or no additional processing. Nevertheless, this process has not been commercialized yet [16]. Theoretically this technique enables the recovery of electrolyte, cathode active material with a little relithiation (whereas about 5% of new lithium are needed), anode and all metallic components present in the battery. The negative point of this technique, despite being still in R&D, is that a very advanced sorting would be necessary at the entry of the process.

Another important point which has already been mentioned above is the need for a high collection rate via an appropriate chain. Currently, the collection rate of used Li-ion batteries is very low or even nonexistent in particular because of their life span (a decade) and their dissemination in various portable applications. Furthermore the current recycling processes and the associated industrial infrastructure cannot assume the future task of recycling of a large flow of spent lithium batteries, since the recycling chains and processes are not designed for this type of batteries. Furthermore, the industrial facilities are not even present in many countries, where the collection and sorting should be done. Thus the recycling of Li-ion batteries requires a robust end-of life infrastructure not existing nowadays.

Moreover, the complexity of the new electrodes composed of several different elements (Co, Ni, Li, Zn, Mn, Fe, etc.) in diverse proportions will certainly be the most important obstacle to be overcome in the future Li-ion recycling chain, especially the focus should be put on the development of appropriate collecting and sorting steps according to their composition. Indeed, the composition and the proportions of these elements will vary significantly. Thus it seems to be very difficult or even impossible to develop a unique process technology for future generations of Li-ion batteries. Currently, several industrial processes for efficient and economical recycling of Li-ion and also Ni-MH batteries exist already. For instance, to recover the various elements of the electrode materials the UMICORE process combines pyrometallurgical and hydrometallurgical steps and the TOXCO process is purely hydrometallurgical. But the question is whether these processes would be adaptable for the future generation of Li-ion batteries and whether these processes would still be cost effective when the proportion of valuable metals in these batteries will decrease.

Up to now only the metals were recovered from the spent batteries. Nevertheless researchers and industrials should focus their efforts on the recycling of other components of the battery too. Especially, the extraction and recycling of electrolytes from Li-ion batteries, which can account for 20% of the weight of the battery, should be studied. Regarding the recycling of metals, much effort should be still done in order to develop efficient recycling technology for the electrodes' active materials as these electrodes will certainly have, in close future, very complex composition as mentioned already above. Indeed different manufacturers will certainly adopt different compositions of their active materials based whether on Li-ion oxides ($LiCoO_2$, $LiMnO_2$, $LiNi_{1/3}Mn_{1/3}Co_{1/3}O_2$...) or on phosphate-based salts ($LiFePO_4$). The organization of the whole recycling chain should also be reflected.

All those end of life processes have their own environmental impacts, and we need to better know the data related to those processes, in order to be able to assess them. But as mentioned in the previous part, to avoid impact transfer between the life cycle phases of lithium ion batteries, it is necessary to focus not only on the recycling technology itself, but to consider the whole life cycle of the product when assessing the environmental impact.

2.2 Life cycle model, analysis of the whole product's life-chain

The researchers and the industrials should not only focus on the end-of-life of the product, but the entire life cycle of the product should be considered. As shown in the figure 9, the sustainability of the battery life cycle depends on four eco-approaches going from the design of the new active material up to the-end-of-life management, whereas the development of an appropriate recycling chain is only the last of them. In order to facilitate the end-of-life treatment of the batteries this problem should be kept in mind from the beginning of the design process (eco-design approach) and the feasibility of recycling of the future active materials should be one of the concerns during the R&D phase. This concern is of the same order of importance as the product improvement from performance point of view. The optimization of the production process is in the heart of eco-production approach. The transport should be minimized, the number of production operation should decrease also, the minimum of harmful products should be used etc. The optimization should also concern the utilization phase (eco-use approach), whereas the life span of the product should be maximized, while the energy consumption should on the contrary be minimized. Nevertheless the previous elements should not be optimized separately, but a global simultaneous optimization process should be preferred in order to avoid the impact transfer. In order to perform this global optimization it is necessary to well define the life cycle model. This model conditions the ability to challenge the design of the whole industrial chain (figure 9).

Insert Figure9

Up to now, most of the impacts downstream in the life cycle of a product comes from the concept & design phase whereas these impacts are traditionally omitted during this phase. Nevertheless the prevention is better than cure and it is much easier to prevent these impacts by taking into account the criteria imposed by regulations and other environmental concerns up from this phase than having to intervene later. Though the decisions made at this early stage will affect the whole life cycle of the product.

Regarding end-of-life phase from an environmental point of view, the objective at this stage is to minimize the impacts of the end of the life of the product as well as of its recycling. At the same time the aim is also to try to recover the maximum of materials that can be reused in the next life-cycle of the product (closed loop). This activity presents several difficulties, mostly originated during the design phase as mention above.

As described in the introduction, one of the major problems in lithium batteries recycling is the difference of technologies (from the chemical point of view) in the input stream, since there is no steady technology yet, and it doesn’t look like that the situation is going to change in the next few years, i.e. importance of LFP, NCM and other technologies on the market.

The efficiency of a recycling process is directly affected by the battery technology sorting before the spent batteries enter this process, as well as the battery configuration, i.e. the way it is assembled, modules and auxiliary components disposition, etc.

Thus it seems today difficult to create a universal – and effective- recycling process, that would be independent from the global industrial chain. Moreover, the design of an effective couple of battery system and its recycling process with low environmental impacts require the assessment of the environmental performances early in the design process. To perform that task, designers would need a model of the whole battery life cycle that supports environmental assessment which does not exists at the present time.

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Figure Captions

Figure 1. Steps of Life Cycle Analysis [4].

Figure 2: Inventory schema where inputs and outputs define a process.

Figure 3: Calculation framework of midpoint indicators from lifecycle inventory.

Figure 4: Impact categories grouped into midpoints, endpoints and single score.

Figure 5: Comparison of the Global Warming Potential impact of two lithium-ion batteries along their life cycle. One uses water as solvent during fabrication and the other battery uses NMP solvent. Source: Zackrisson et al. [7].

Figure 6: Use phase analysis results (global warming, photochemical smog, eutrophication, acidification and ozone depletion) for a 10 kWh lithium-ion battery, used in a Pluged-In Hybrid Electric Vehicle (PHEV). Source: Zackrisson et al. [7].

Figure 7: LCA results (global warming, photochemical smog, eutrophication, acidification and ozone depletion) for a 10 kWh lithium-ion battery, used in a Pluged-In Hybrid Electric Vehicle (PHEV). Source: Zackrisson et al. [7]

Figure 8: Hydro-metallurgy versus pyro-metallurgy processes.

Figure 9: Eco-design of the whole battery life chain.

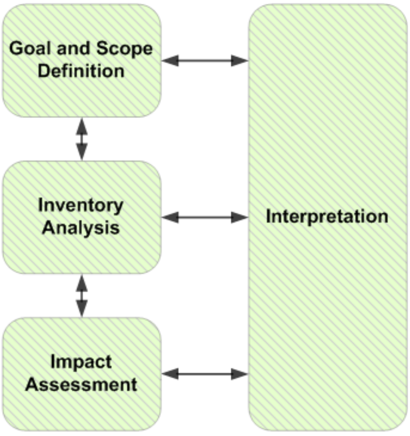


Figure 1. Steps of Life Cycle Assessment [4].

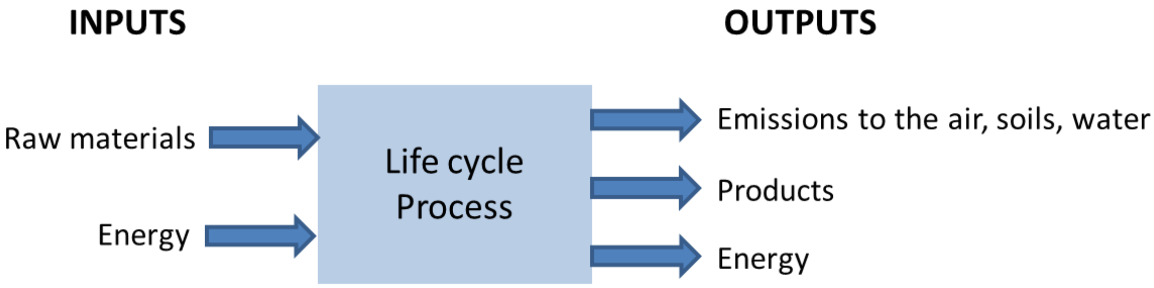


Figure 2: Inventory schema where inputs and outputs define a process

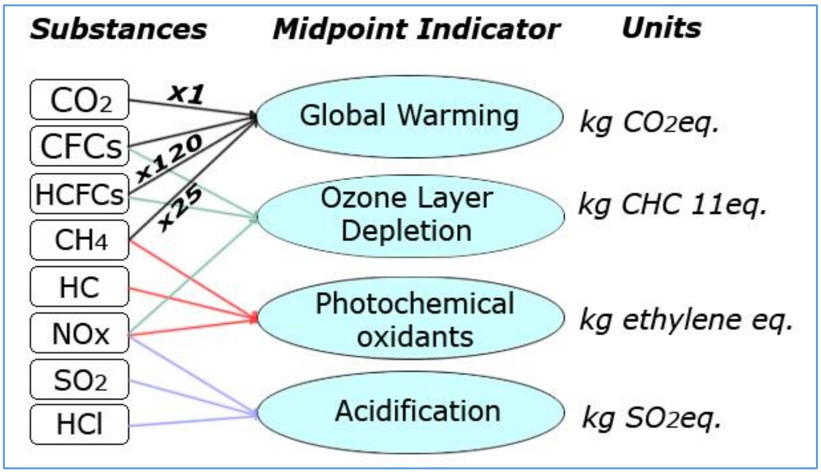
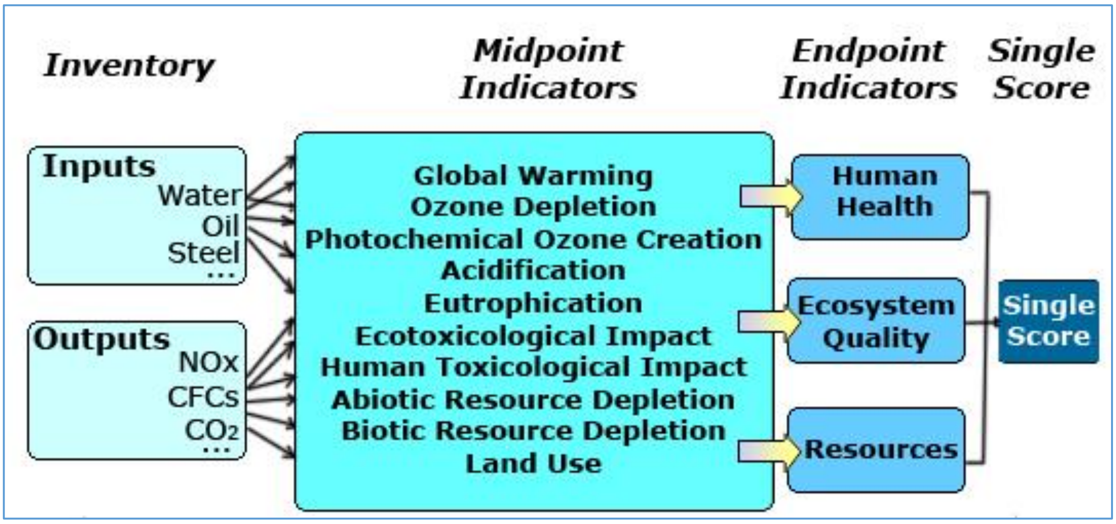


Figure 3: Calculation framework of midpoint indicators from lifecycle inventory



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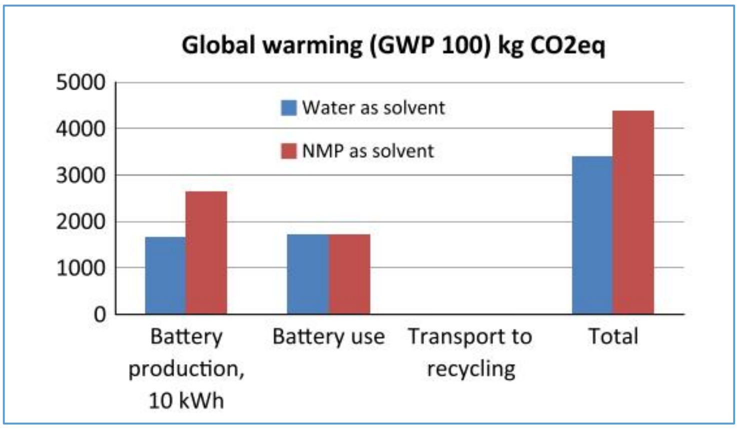


Figure 5: Comparison of the Global Warming Potential impact of two lithium-ion batteries along their life cycle. One uses water as solvent during fabrication and the other battery uses NMP solvent. Source: Zackrisson et al. [7]

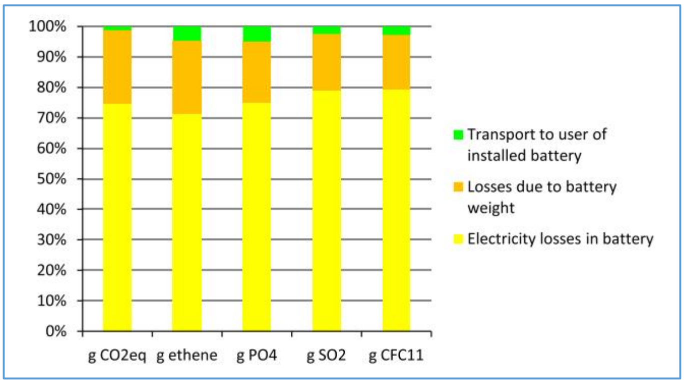
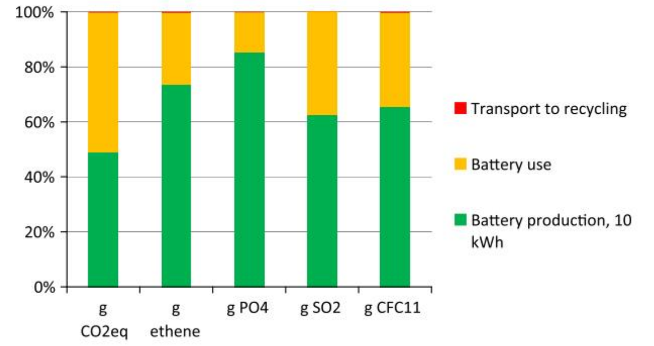


Figure 6: Use phase analysis results (global warming, photochemical smog, eutrophication, acidification and ozone depletion) for a 10 kWh lithium-ion battery, used in a Pluged-In Hybrid Electric Vehicle (PHEV). Source: Zackrisson et al. [7]



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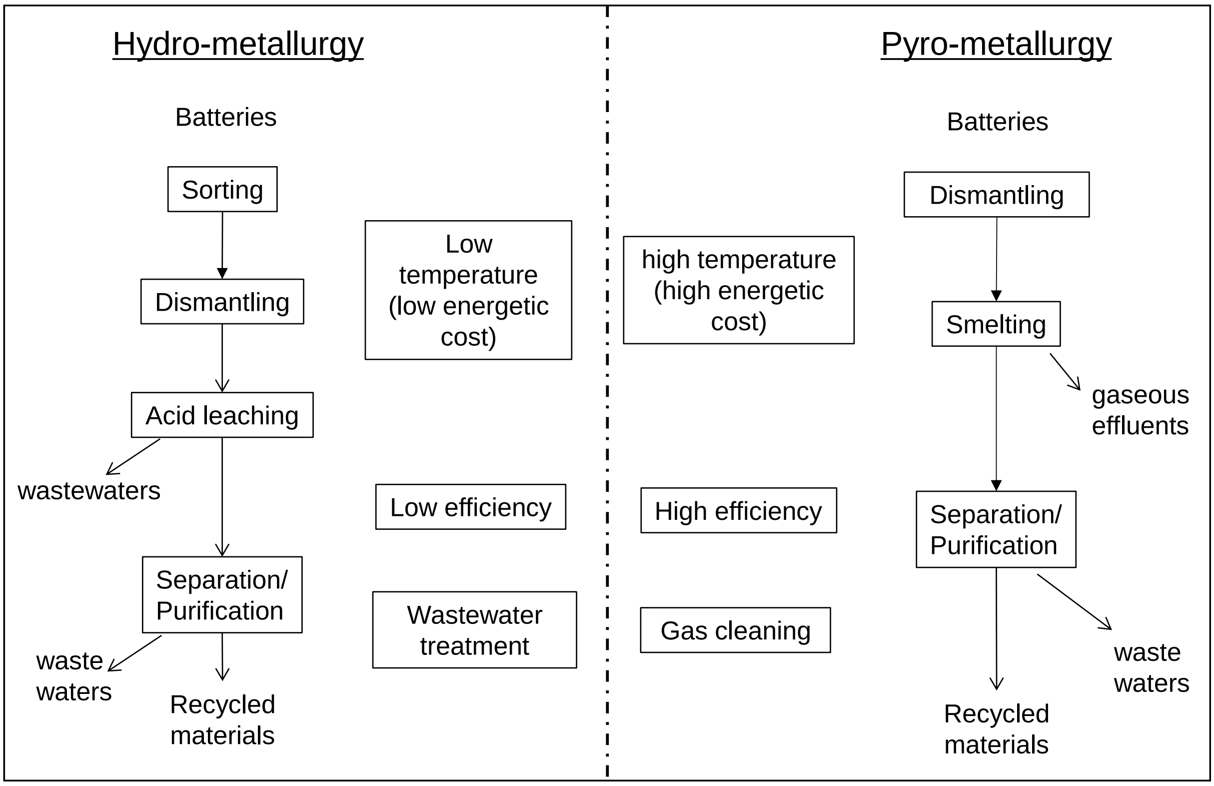

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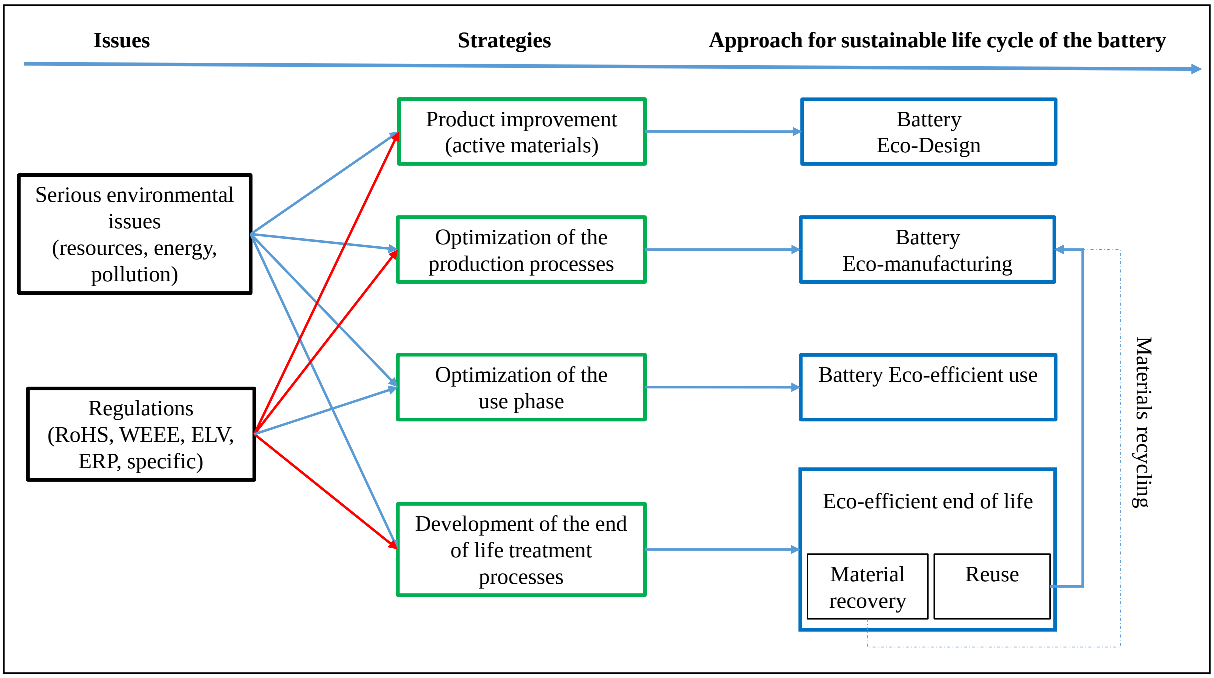

Figure 9: Eco-design of the whole battery life chain.

Table captions

Table 1 : Comparison of discussed studies.

Table 2 : LCA stages taken into account by the different authors.

Table 3 : Environmental impact categories selected by authors.

Table 4. Calculation Methods used by each researcher.

Table 5. Recycling threshold depending on battery technology according to EU 2006/66/CE directive [14].

Table 1: Comparison of discussed studies.

Author	Functional Unit	Cathode Chemistry*	LC phases considered	Key characteristic
EPA [5]	km travelled by vehicle	LMO, LFP, NCM	All	10 years lifetime (1 battery per vehicle)
Majeau-Bettez et al. [6]	50 MJ	NCM, LFP	Extraction of minerals and Fabrication	No differences between PHEV** and EV** batteries.
Zackrisson et al. [7]	10 kWh	LFP	All	Battery capacity: 93 Wh/kg, 3000 cycles at 80% depth of discharge.
Dunn et al. [8]	kg of battery	LMO	*Cradle-to-Gate* but no use phase	Global Warming Potential and Energy only
Ellingsen et al. [10]	One battery pack	NCM	Extraction of minerals and Fabrication	-
Notter et al. [9]	km Travelled by vehicle	LMO	All (but landfilling)	-

*LMO : lithium manganese oxide, LFP : Lithium Iron Phosphate, NMC : Nickel Manganese Cobalt oxide

** PHEV: Plug-In Hybrid Vehicle. EV: Electric Vehicle.

Table 2: LCA stages taken into account by the different authors

	Raw materials extraction	***Active material processing***	***Battery manufacturing***	***Additional components***	***Use phase***	***End of life***
EPA [5]	Inside boundaries	Inside boundaries	Inside boundaries	Inside boundaries	Inside boundaries	Inside boundaries
Majeau-Bettez et al. [6]	Not included in the study	Inside boundaries	Inside boundaries	Inside boundaries	Inside boundaries	Not included in the study
Zackrisson et al. [7]	Inside boundaries	Inside boundaries	Inside boundaries	Inside boundaries	Inside boundaries	Not included in the study
Dunn et al. [8]	Inside boundaries	Inside boundaries	Inside boundaries	Inside boundaries	Inside boundaries	Inside boundaries
Notter et al. [9]	Inside boundaries	Inside boundaries	Inside boundaries	Inside boundaries	Inside boundaries	Inside boundaries
Ellingsen et al. [10]	Inside boundaries	Inside boundaries	Inside boundaries	Inside boundaries	Not included in the study	Not included in the study

Table 3: Environmental impact categories selected by authors

Author	Impact Categories
EPA [5]	Energy, ADP, GWP, Acidification, Eutrophication, ODP, POP, Ecological Toxicity, HTP, Land occupation, cancer & non cancer hazard
Majeau-Bettez et al. [6]	Energy, GWP, FDP, FETP, FEP, HTP , METP, MEP, MDP, ODP, PMFP, TAP, TETP
Zackrisson et al. [7]	Energy, GWP, Acidification, ODP, Photochemical smog, Eutrophication
Dunn et al. [8]	Energy, GWP
Notter et al. [9]	ADP, nonrenewable cumulated energy demand, GWP
Ellingsen et al. [10]	Energy, GWP, FDP, ODP, POFP, PMFP, TAP, FEP, MEP, FETP, METP, TETP, HTP, MDP

Impact categories: abiotic depletion potential (ADP) ***kg Sb eq*****, photochemical oxidation potential(POP)** ***kg O_3 eq*****, global warming potential (GWP)** ***kg CO_2 eq*****, fossil depletion potential (FDP)** ***kg oil eq*****, ozone depletion potential (ODP)** ***kg CFC 11 eq*****, photo oxidation formation potential (POFP)** ***kg nonmethane volatile organic carbon*****, particulate matter formation potential (PMFP)** ***kg PM10 eq*****, terrestrial acidification potential (TAP)** ***kg SO_2 eq*****, freshwater eutrophication potential (FEP)** ***kg P eq*****, marine eutrophication potential (MEP)** ***kg N-eq*****, freshwater toxicity potential (FETP)** ***kg 1,4-dichlorobenzene eq*****, marine toxicity potential (METP)** ***kg 1,4-DCB eq*****, terrestrial eutrophication potential (TETP)** ***kg 1,4-DCB eq*****, human toxicity potential (HTP)** ***kg 1,4-DCB eq*****, and metal depletion potential (MDP)** ***kg Fe eq*****.**

Table 4. Calculation Methods used by each researcher.

Author	Calculation Method
EPA [5]	Several methods depending on the indicator
Majeau-Bettez et al. [6]	ReCiPe [13]
Zackrisson et al. [7]	Several methods depending on the indicator
Dunn et al. [8]	BatPaC[1], for mass inventories. GREET model[2]
Notter et al. [9]	EcoIndicator EI99 H/A [11] CML [12]
Ellingsen et al. [10]	ReCiPe [13]

[1] http://www.cse.anl.gov/batpac/index.html
[2] https://greet.es.anl.gov/

Table 5. Recycling threshold depending on battery technology according to EU 2006/66/CE directive [14].

Type	***Recycling***	
	Rate (% w)*	Heavy metals (% w)**
Lead acid	65%	around 100%
Ni/Cd	75%	
Other chemistry (Ni-MH, Li-ion, …)	50%	-

(* compulsory, ** recommended).