

Case Report

Future Portable Li-Ion Cells' Recycling Challenges in Poland

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Abstract: The paper presents the market of portable lithium-ion batteries in the European Union (EU) with particular emphasis on the stream of used Li-ion cells in Poland by 2030. In addition, the article draws attention to the fact that, despite a decade of efforts in Poland, it has not been possible to create an effective management system for waste batteries and accumulators that would include waste management (collection and selective sorting), waste disposal (a properly selected mechanical method) and component recovery technology for reuse (pyrometallurgical and/or hydrometallurgical methods). This paper also brings attention to the fact that this EU country with 38 million people does not have in its area a recycling process for used cells of the first type of zinc-carbon, zinc-manganese or zinc-air, as well as the secondary type of nickel-hydride and lithium-ion, which in the stream of chemical waste energy sources will be growing from year to year.

Keywords: spent batteries and accumulators; Li-ion cells; legislation; recycling

1. Introduction

Lithium-ion chemical energy sources (Li-ion) dominate the market for secondary type batteries (accumulators); almost all mobile phones and laptops are powered by lithium cells [1,2]. In addition, the growing market for electric and hybrid cars [3] is a new industry generating demand for Li-ion batteries. Lithium-ion batteries contain several of valuable metals such as cobalt, copper, lithium, nickel, manganese, aluminium and iron [4–6]. Cobalt is one of the less common metals in the Earth's crust, hence its market value is high at 80,491\$/MT [7], and its recovery is profitable [8]. Lithium recovery is essential for the development of electric car production [9]; the current price is 16,500\$/MT [7]. Mass production of vehicles powered by lithium-ion batteries will increase demand for lithium, so its price will increase and recovery will be profitable [10–12]. Another argument in favour of recycling batteries is the need to protect the environment from pollution by heavy metals or complex organic substances contained in Li-ion batteries [13,14].

It is also very important that, despite a decade of efforts in Poland, it has not been possible to create an effective management system for waste batteries and accumulators that should include waste management (collection and selective sorting), waste disposal (a properly selected mechanical method) and component recovery technology for reuse (pyrometallurgical and/or hydrometallurgical methods). The fact that this European Union country with a population of 38 million does not have in its area a recycling process for used cells of the first type of zinc-carbon, zinc-manganese or zinc-air, as well as the secondary type of nickel-hydride and lithium-ion, which in the stream of chemical waste energy sources will be growing from year to year, means there is a potential benefit from a future solution.

2. Market for Portable Batteries and Accumulators in the European Union (EU) and Poland

The total mass of portable batteries and accumulators placed on the EU market between 2009 and 2017 is estimated at 1,921,000 MT and collected at 694,000 MT [15]. Data for 2017, not yet collected from all member states of the EU, show that in 2016 215,000 MT portable cells were introduced and 94,000 MT (~43.72%) were collected. The largest mass of portable chemical energy sources was introduced to the EU market by Germany 45,511 MT (~21.2%), the UK 38,659 MT (~18.0%), France 29,491 MT (~13.7%) and Italy 25,197 MT (~11.7%). Poland is ranked fifth with a weight of 12,585 MT, which accounted for about 5.9% of all portable cells on the European market two years ago. Unfortunately, there are no data available on portable lithium-ion batteries and accumulators passing through the EU market. According to data published on the Accurec Recycling GmbH website [16], around 70,000 MT of Li-ion cells for different applications will be introduced to the EU market in 2020, while only slightly more than 8000 MT of this type of waste will be recycled. Nevertheless, it is estimated that the global market for lithium cells will be worth USD 93.1 billion in 2025 [17] and that around 50,000 MT of such waste will be recycled in the member states by this year [18].

In Poland, pursuant to Article 72 of the Batteries and Accumulators Act 2009 [19], the Chief Inspector of Environmental Protection prepares an annual report on the state of management of batteries and accumulators and their waste. The report contains data on the number of companies placing batteries and accumulators on the market as well as on the number of plants collecting and processing waste chemical energy sources. The report includes information on the quantity and weight of batteries placed on the market as well as collected and treated battery waste, distinguishing between three categories: portable, automotive and industrial batteries and accumulators. In the register of chemical energy sources introduced to the Polish market, data on the mass and quantity of batteries and accumulators of the following types are collected separately: nickel-cadmium (Ni-Cd), lead-acid (Pb-Acid), button cells with mercury, button cells without mercury, zinc-carbon (Zn-C), zinc-manganese (Zn-Mn) and zinc-air (Zn-O₂). Batteries and accumulators that do not belong to any of these groups together form the last, sixth group—other batteries and accumulators. The register of collected waste batteries contains separate data on the quantity and weight of nickel-cadmium and lead-acid batteries and accumulators; all other types of batteries are counted together. In Poland, data on the number of lithium-ion batteries introduced to the market and collected are not gathered. In the annual reports of the Chief Inspector of Environmental Protection, the stream of the accumulators discussed forms part of the “other batteries and accumulators” group, which also includes silver, lithium and nickel-hydrogen accumulators.

According to the latest available report of the Chief Inspectorate of Environmental Protection of 2018, at the end of 2017 there were 3648 registered entrepreneurs placing batteries or accumulators on the market (1035 entrepreneurs placing batteries and accumulators on the market and 2613 entrepreneurs placing batteries or accumulators together with electrical and electronic equipment on the market) and 25 entrepreneurs operating in the field of waste batteries and accumulators processing. In total, 134,951.6 MT of batteries and accumulators were placed on the market in 2017, including 13,269.9 MT of portable batteries and accumulators, 30,306.5 MT of industrial batteries and accumulators, and 91,375.3 MT of automotive batteries and accumulators. Detailed data on the types of chemical energy sources placed on the market are broken down into three categories: portable, automotive and industrial. Both automotive and industrial batteries are mostly lead-acid batteries. The data collected in the reports of the Chief Inspectorate of Environmental Protection [20–27] show that they account for over 99% of automotive batteries and between 90% and 98% of industrial batteries. The largest number of lithium-ion batteries is used in mobile phones and personal computers and belongs to the category of portable batteries and accumulators [28].

Table 1 shows the number of portable batteries and accumulators introduced to the Polish market according to the data contained in the reports on the functioning of battery and accumulator management for the years 2010–2017 [20–27].

Table 1. Mass of batteries and accumulators introduced to Polish market in 2010–2017, in MT [20–27].

Year	2010	2011	2012	2013	2014	2015	2016	2017
Zn-Mn, Zn-C, Zn-O ₂	5976.3	5451.3	5715.3	6515.3	7371.5	7343.4	7540.4	8259.9
Ni-Cd	662.9	660.1	489.4	449.4	606.2	375.2	390.5	212.6
Pb	348.9	369.5	296.3	413.6	409.7	416.5	281.7	413.4
Button-type batteries and accumulators without Hg	179.3	153.3	113.9	143.6	141.8	186.9	232.0	279.6
Button-type batteries and accumulators from Hg	2.4	3.6	5.6	22.5	8.27	7.8	16.3	8.4
Other batteries and accumulators	2696.5	3133.4	3978.6	3719.4	3261.3	3875.5	4124.3	4096.0

Table 1 shows that in 2017 more than 13,000 tons of portable batteries and accumulators were introduced to the market; 62.2% of this amount were zinc-carbon, alkaline and zinc-air batteries. The other specified types, i.e., button cell batteries and nickel-cadmium and lead-acid batteries, accounted for a small share of 6.9%. Whereas 30.9%, in 2017, i.e., almost 4.1 thousand tonnes, are other cells, i.e., silver, nickel-hydride, lithium and lithium-ion cells. The use of silver and lithium cells is relatively small, while nickel and hydrogen batteries are used mainly in hybrid cars and are currently being replaced by lithium ion batteries.

3. Li-Ion Battery and Accumulator Stream in Poland

In order to determine the size of the waste stream of Li-ion batteries, the necessary information is the level of effectiveness of their collection. The Chief Inspectorate of Environmental Protection [20–27] reports contain data on the amount of portable batteries and accumulators collected. Collection rates for waste batteries are set by the Directive on batteries and accumulators [29] and are: 25% from 2012 to 45% from 2016. In Poland, the required collection rates are specified in the Ordinance of 3 December 2009 on annual collection rates of waste portable batteries and waste portable accumulators [30], which in subsequent years were as follows: 2010—18%, 2011—22%, 2012—25%, 2013—30%, 2014—35%, 2015—40%, 2016 and 2017—45%. The required levels of collection of used energy sources were achieved by 2013 (2010—18.00%, 2011—22.72%, 2012—29.10%). In 2014 and 2015 to achieve an appropriate level of collection of this type of waste, approximately 2.0% (2014—33.06%, 2015—38.35%) were missing. The last three years have seen a significant increase in the collection of waste cells and so in 2015 54.92%, in 2016 78.14% and 2017 65.74% of waste portable batteries and accumulators were collected from the market. The lack of detailed information on the collection efficiency of waste Li-ion batteries makes it clear that, in order to estimate the amount of the waste stream, the collection rate of Li-ion batteries is equal to the collection rate of all portable batteries and accumulators. Assuming also that lithium-ion batteries constitute 80% of batteries and accumulators other than zinc-carbon, alkaline, zinc-air, nickel-cadmium, lead-acid and button batteries and accumulators, it is possible to calculate the stream of used Li-ion cells in the years 2010–2017 (Figure 1). Calculations show that from 2010 to 2017 the mass of Li-ion batteries introduced to the Polish market increased by 34.17% from 2157.24 MT to 3276.79 MT. However, the total stream of collected waste lithium cells in the last seven years could amount to as much as 10 076.15 MT and it should be noted that Poland is a country where there is no single technology for processing waste batteries and accumulators. Processing methods of portable batteries concern only used cells of the first type of Zn-C, Zn-Mn and Zn-O₂ cells and end with mechanical processing and separation of three ferromagnetic, diamagnetic and paramagnetic material fractions for management [31]. Based on the value of masses of Li-ion batteries and accumulators introduced into and collected from the Polish market, it can be concluded that in 2020 these streams may amount to 4361.40 MT and 1962.63 MT respectively. In 2030, the volume of these streams may increase 2.6 times to 11,312.36 MT and 5090.56 MT in relation to 2020. These forecasts are consistent with the data presented in the works of Rogulski and Czerwiński [32] and Rogulski and Dłubak [33],

in which the authors presented a detailed analysis of the portable batteries market in Europe in terms of the management of electrochemical energy sources.

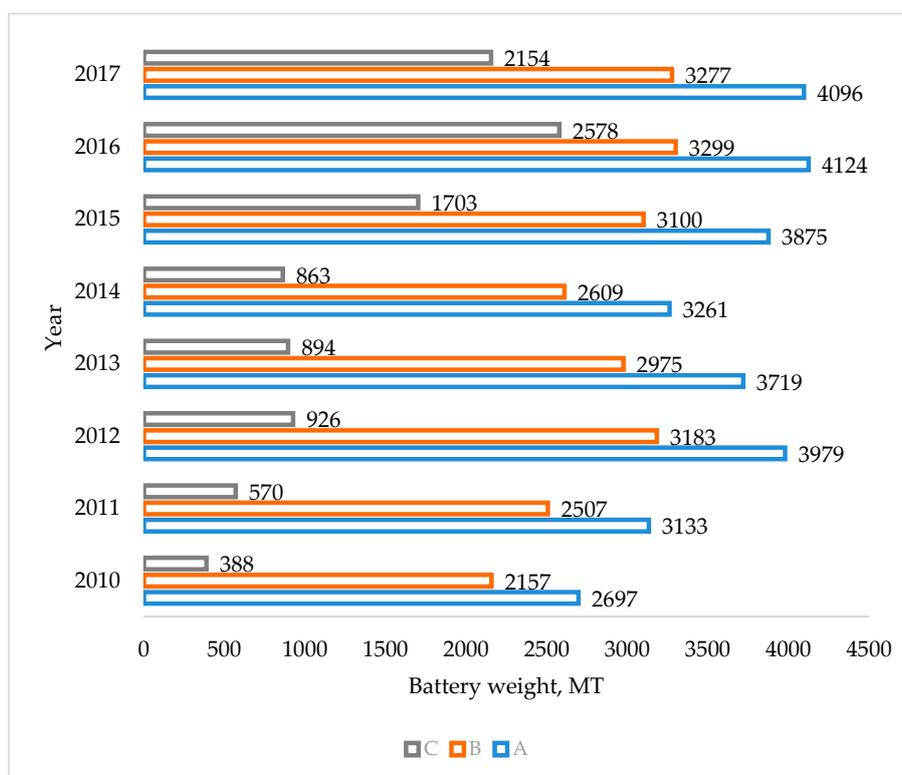


Figure 1. Estimated volume of Li-ion battery and accumulator streams in Poland in 2010–2017: A—weight of other batteries and accumulators placed on the market; B—weight of Li-ion batteries and accumulators placed on the market; C—mass of accumulated used Li-ion batteries and accumulators, in MT.

4. Recycling of Used Li-Ion Batteries and Accumulators in Poland

The reference system for the recycling of used batteries and accumulators should be based on three complementary and successive unit processes. The first of these is a mechanical treatment most often used for large cells (industrial type) and as a preliminary operation in most processing technologies. Separation processes involve the mechanical loosening of the structure (body) of the battery and separation of components with characteristic physical properties (density, size, magnetic properties). These activities are usually simple and cheaper than other processes, and for that reason they should be used to prepare the material stream for further processing [31–34]. The second main processes are pyrometallurgical and/or hydrometallurgical processes. Pyrometallurgical methods rely on the recovery of materials (in particular metals) by carrying them out at sufficiently high temperatures to specific condensed phases (including a metallic alloy) or to the gas phase with subsequent condensation. In general, these methods are more appropriate for phases rich in recoverable components, possibly concentrating at elevated temperatures in the gas phase (this applies to e.g., mercury removal, cadmium or zinc extraction). However, it should be remembered that this division is arbitrary and is not suitable for strict chemical or technological considerations. With regard to batteries, these processes can be carried out both in a traditional way, i.e., using the oxidation-reduction equilibria of the HCO system (hydrogen, carbon, oxygen), and in an extended manner, which is characteristic of advanced chemical metallurgy, where for example chlorination processes are utilized. The advantage of pyrometallurgical methods is the possibility of recycling various types of cells, including those containing various organic materials [35]. In contrast, hydrometallurgical methods usually rely on acid or alkaline leaching of properly prepared battery waste (after machining processes). They are followed by a series of

physicochemical operations that lead to the separation and concentration of valuable or burdensome components between the respective phases, up to commercial products and semi-finished products for separate technological processes (pyrometallurgical or hydrometallurgical) or waste. It is believed that hydrometallurgical processes are less energy-consuming than pyrometallurgical ones, but the waste from them is more burdensome. The advantage of hydrometallurgical processes is also that they allow in most cases the processing of a mixture of different types of batteries simultaneously [36–38]. The third stage of recycling used cells should be the processes of managing all post-process waste in such a way that they are not harmful to the environment.

In 2017, there were 25 registered companies operating in the field of processing waste batteries and accumulators in Poland [27]. However, in reality only a few companies conduct raw material recovery from waste cells—Table 2 [39–47]. Processing of used zinc–carbon, zinc–manganese and zinc–air batteries is mainly mechanical, which results in creation of ferromagnetic fractions, diamagnetic fraction and paramagnetic fraction. The ferromagnetic fraction consisting of metals such as iron, chromium and nickel is a secondary raw material for steel works. The diamagnetic fraction in which plastics and paper accumulate, when mixed with sawdust and used cleaning cloth, is used as a substrate for the production of alternative fuel, the recipients of which are cement plants or combined heat and power plants. The paramagnetic fraction is mainly dominated by graphite and non-ferrous metals with the remainder of the other two fractions [48]. However, zinc and manganese contained in the last fraction that could be obtained by pyrometallurgical [49] or hydrometallurgical [50] methods are not recovered. The lack of a complete domestic technology for recycling used cells of the first type means that secondary raw materials containing valuable metals are resold to foreign companies based in Finland, Germany or Slovenia [27]. Only lead-acid cells are recycled in high-efficiency pyrometallurgical installations. Baterpol S.A. and Orzeł Biały S.A. Process Pb-acid cells by thermal methods to obtain lead alloys and polypropylene, which are again used in the production of car batteries [45,46].

Lithium-ion cells are not processed in any domestic installation, they are probably sorted out of the entire stream of used batteries and accumulators, collected and then resold at a price of 300 to 1000\$/MT [51] to foreign companies having installations for their recycling. Therefore, assuming the average price of 600\$/MT of used lithium-ion batteries collected in Poland, it can be calculated that the market value of the discussed waste in 2017 was approximately 1.30 million USD and in 2030 reached the value of 3.05 million USD. It seems that the estimated amounts, apart from the costs of designing and constructing the installation and its current maintenance, could give the potential investor real profits in the next 10 years. Of course, press reports show that several companies alone or in cooperation with research institutions are planning or are implementing research projects related to the creation of recycling technologies for used Li-ion batteries and accumulators [52–54]. However, it is difficult to deduce from this information in what time perspective a complete recycling installation could be built in Poland.

Table 2. Technologies for processing used batteries and accumulators in Poland [39–47].

Name of Technology	Recycling Method	Type of Remanufactured Battery	Recovered Metals and Compounds
BatEko	Mechanical processing	Zn–C	— ferromagnetic fraction
		Zn–Mn	— diamagnetic fraction
		Zn–Air	— paramagnetic fraction
Grupa Eneris	Mechanical processing	Zn–C	— metallic Zn
		Zn–Mn	— brass
		Zn–Air	— steel
			— RDF (foil and paper)

Table 2. Cont.

Name of Technology	Recycling Method	Type of Remanufactured Battery	Recovered Metals and Compounds
MB Recycling Sp. z o. o.	Mechanical processing	Zn-C Zn-Mn Zn-Air	— ferromagnetic fraction — diamagnetic fraction — paramagnetic fraction
Biosystem S. A.	Mechanical processing	Zn-C Zn-Mn Zn-Air Ni-Cd	— ferromagnetic fraction — diamagnetic fraction — paramagnetic fraction
MarCo Ltd. Sp. z o. o.	Mechanical processing	Ni-Cd	— iron-nickel electrodes — iron-cadmium electrodes — steel scrap — potassium lye — plastic waste
Eco Harpoon Recycling Sp. z o.o.	Mechanical processing	Zn-C Zn-Mn Zn-Air Ni-Cd Batteries containing mercury	— ferromagnetic fraction — diamagnetic fraction — paramagnetic fraction — mercury — cadmium — ferrous and non-ferrous metals
Baterpol S.A.	Pyrometallurgical	Pb-acid	— alloys Pb — solid Na ₂ SO ₄ — polypropylene
Orzeł Biały S.A.	Pyrometallurgical	Pb-acid	— alloys Pb — polypropylene pellets
ZM Silesia S.A.	Pyrometallurgical	Ni-Cd	— metallic Cd — ferro-alloys containing Ni

5. Conclusions

The above quantitative and qualitative analysis of the Polish market for batteries and accumulators as well as waste batteries and accumulators shows that in the next 10 years in the country the stream of discussed waste will change dynamically. In addition to the used cells of the first type, the number of which on the market is still high, one should expect an additional significant growth in number of cells of the secondary type, especially the lithium-ion type. That is why it is so important that the current system of managing chemical energy sources undergo versification and modification taking into account the changes associated with the technology of producing new cells, which in the near future will become a valuable source of secondary raw materials. Meanwhile despite an increase in the level of metal recovery from used chemical energy sources, the amount of batteries placed on the market is growing so rapidly that the problem of recycling of secondary type cells is constantly worsening. This is influenced in particular by the development of personal device technology, which mostly uses lithium-ion cells. Despite a number of publications and reports raising the high level of risk related to pollution of the environment by poorly managed cells containing highly harmful substances, the first overriding problem limiting the effectiveness of the recovery of raw materials from lithium-ion cells is low social awareness resulting in a small stream of selectively collected waste batteries. As a result,

only every tenth cell is recycled, which has a direct impact on the high technological costs associated with the recovery of metals from smaller waste material stream. The task of states and properly appointed institutions should be to take care not only of the interests of the present but also future generations, who may have major problems with maintaining stable development and rational raw material management. Greater efforts should be made to improve public education and raise consumer awareness of the need to return raw materials for re-use, not only through regulations imposing minimum recycling rates, which usually involve higher costs, but also through improvements in the raw material situation. Also important is improvement of social awareness, especially in developed countries, where the number of devices containing cells introduced to the market is the first barrier to achieving rational levels of recovery of raw materials from electronic devices. Technologies for recovering metals from battery cells are slowly surpassing the technologies used to produce them. This is a signal that they should have a real impact on battery manufacturers to look for solutions that allow easier recovery of components and materials used in their production. There is now a clear tendency to optimize the production of cells, not only lithium-ion cells, in terms of their cost of production in relation to their efficiency without considering recovery and reuse. As a result, the resulting batteries are not prepared for proper treatment after use, and the costs associated with the recovery of materials are increasing.

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