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## Collection and recycling of spent nickel and lithium batteries and accumulators in Poland

### Zbiórka i recykling zużytych baterii i akumulatorów niklowych i litowych w Polsce

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#### **Abstract**

The European Union has established strict requirements for the recycling of spent batteries and accumulators. In Poland, this waste is currently recycled pyrometallurgically solely for Cd, Zn, Pb and ferronickel recovery. However, waste cells (Ni-Cd, Ni-MH, Li-ion) represent a source of strategic metals (Ni, Co, Li). This paper shows an analysis of the market and applications of nickel and Li-ion batteries as well as current state of domestic collection and recycling of spent cells.

**Keywords:** batteries, cadmium, lithium, nickel, recycling

#### **Streszczenie**

Unia Europejska stawia państwom członkowskim wysokie wymagania dotyczące recyklingu zużytych baterii i akumulatorów. W Polsce z odpadów tego typu odzyskuje się wyłącznie Cd, Zn, Pb i żelazonikiel w procesach pirometalurgicznych. Jednak zużyte ogniwa (Ni-Cd, Ni-MH, Li-ion) mogą stanowić źródło metali strategicznych (Ni, Co, Li). W artykule przedstawiono analizę rynku i zastosowań baterii i akumulatorów niklowych i Li-ion oraz omówiono aktualny stan krajowej zbiórki i recyklingu zużytych ogniw.

**Słowa kluczowe:** baterie, kadm, lit, nikiel, recykling

## 1. Introduction

Domestic demand for various metals force us to search new sources of metals [1]. Since we produce only some of them (mainly copper, silver, rhenium, zinc, cadmium, and lead), it is necessary to import the rest. In recent years, there has been an increased interest

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in the recovery of metals from waste materials in Poland [2]. Recycling companies insist mainly the treatment of WEEE despite the fact that there are also opportunities to recover valuable elements from other sources. The latter concerns spent batteries and accumulators (portable, automotive, and industrial). National recycling plants specialize in pyrometallurgical processing of only certain groups of spent cells (mainly zinc, lead-acid, and Ni-Cd) for the recovery of zinc, lead, cadmium, and ferronickel [3, 4]. So far, no domestic industrial installation for the recovery of nickel, cobalt, or lithium from spent cells has been launched. Waste batteries and accumulators are also sent abroad (e.g., to France, Finland, Germany, and Sweden), where they are treated for the selective recovery of metals [5, 6].

Analysis of the current trends in global markets shows that the development and competition of the potential economic giants such as China and India can affect the prices of metals for a long period of time. Furthermore, some limitations in the easily accessible ore deposits are predicted. There is also the risk of a lack of supply of certain raw materials associated with the domination of global production by some countries; for example, nearly 45% of cobalt is supplied by the Democratic Republic of Congo, approx. 17% of nickel by Russia, and 44% of lithium by Chile [7]. All indicate that the recovery of metals from a variety of waste products will soon play a very serious role in the world production of such metals. Such a trend is already called “urban mining”.

## **2. Spent batteries and accumulators as a source of metals**

Batteries and accumulators are sources of electrical current for a wide range of devices, both in household and industrial applications. The EU data shows [8] that currently portable electric devices are powered mainly by Li-ion (60%) and Ni-Cd (34%) cells, while Ni-MH batteries account for a small proportion (6%). The natural tendency in battery sales, however, is replacing Ni-Cd batteries with Li-ion and Ni-MH batteries. It is estimated that only the cordless-tool market in the EU will increase by 5% annually until 2020, with a simultaneous decrease in the Ni-Cd battery supply, especially since the European Parliament has banned the use of Ni-Cd batteries in such applications starting in December 2016. In the coming years, despite the increase in the number of batteries and accumulators placed on the market, only a slight upward trend in the number of waste batteries and accumulators may be noted due to significant improvements in the quality of the cells and prolongation of their life span.

The Polish market of electrical and electronic devices is also rapidly growing [2]. This is accompanied by the growing amount of spent portable batteries and accumulators collected every year; thus, the problem of the waste management.

The above consideration shows that there is a need for the development of cheap and effective domestic technologies for the recovery of metals (nickel, cobalt, REE) from discarded cells.

Used batteries and accumulators consist of materials of various compositions. They contain (on average):

- Ni-Cd [9]: Cd 12–15%, 18–30% Ni, and 0.4–2% Co. These elements are in the form of metals (Ni, Cd) or hydroxides ( $\text{NiOOH}$ ,  $\text{Ni(OH)}_2$ ,  $\text{Cd(OH)}_2$ ,  $\text{Co(OH)}_2$ ). The cells are enclosed in a steel housing. The space between the electrodes (cadmium anode, nickel cathode) is filled with an aqueous electrolyte solution containing approx. 30% KOH, sometimes with an admixture of NaOH (below 4%) or LiOH (0.5–2%). To avoid a short circuit, the anode spaces are separated from the cathode with separators made of cellulose derivatives.
- Ni-MH [10]: 23–60% Ni, 4–8% Co, and 20% lanthanides (La, Ce, Pr, Nd), small amounts of Mn, Zn, and Al. They may also include Ti, Cr, V, and Zr. Metals are mainly in the form of  $\text{AB}_2$  and  $\text{AB}_5$  alloys. In the first case, the anode usually consists of intermetallic compounds  $\text{MnNi}_5$  and  $\text{LaNi}_5$  (the latter converts into  $\text{LaNi}_5\text{H}_6$  during charging). The  $\text{AB}_2$  alloy is represented commonly by  $(\text{Ti}_{2-x}\text{Zr}_x\text{V}_{4-y}\text{Ni}_y)_{1-z}\text{Cr}_z$ , where  $x = 0-1.5$ ;  $y = 0.6-3.5$ ;  $z < 2$ . The Ni-Mn-Co alloy can be also used. The cathode active material is mainly  $\text{Ni(OH)}_2$  deposited on a porous nickel substrate. The electrolyte is KOH (10–15%). The anode is separated from the cathode with a synthetic porous membrane. Steel housing represents approx. 15–25% of the cell mass.
- Li-ion (secondary cell) [11]: metals (up to 20% Co, 21% Ni and 5–7% Li), organic compounds (15%), and plastics (7%). Cathode material is typically  $\text{LiCoO}_2$ , but  $\text{LiMn}_2\text{O}_4$ ,  $\text{LiNiO}_2$ ,  $\text{LiMn}_x\text{Ni}_y\text{Co}_z\text{O}_2$ , and  $\text{LiNi}_x\text{Co}_y\text{Al}_z\text{O}_2$  or  $\text{LiFePO}_4$  are also used. Cathode material is coated on aluminum foil by means of a polymeric adhesive material (PVDF), while the graphite anode active material is deposited on copper foil. The electrolyte consists of organic compounds (e.g., propylene carbonate, ethylene carbonate, DMSO), in which lithium salts are dissolved (usually  $\text{LiPF}_4$ , but also  $\text{LiBF}_4$ ,  $\text{LiClO}_4$ ,  $\text{LiCF}_3\text{SO}_3$ ,  $\text{Li}(\text{SO}_2\text{CF}_3)_2$ ). The cells are sealed in a steel or (more commonly) aluminum housing. Recycling of the spent Li-ion cells is conducted mainly for the recovery of cobalt and lithium; but currently, the most cost-effective is the regeneration of  $\text{LiCoO}_2$  cathodes. The treatment of used batteries should be approached with a particular caution because of the possibility of fire and explosion due to the rapid oxidation of metallic lithium (produced at the anode due to overcharging) in the presence of air or water vapor.

Cadmium is well-known as an element with toxic properties. This resulted in a reduction of production and certain applications of the metal and its compounds in the world (e.g., Ni-Cd batteries). However, it is predicted that cadmium will be still used for the production of lighting, alarm systems, medical equipment, dyes, or modern photovoltaic cells. Already launched rechargeable Ni-Cd cells and the collection of spent cells will be a significant source of cadmium recovery in Poland. The available data [1, 7] shows that the current domestic production of cadmium fully meets the demand

for the metal in Poland, which is also associated with the export to the Belgian and Chinese markets.

In contrast to cadmium, nickel is used for the manufacture not only the Ni-Cd batteries but also Ni-MH cells at an increasing extent. They are applied in telecommunications, emergency lighting systems, and hybrid cars [12]. In 2014, more than 6.3 million Ni-Cd batteries were sold in Poland, [5] (not counting other kinds of nickel-bearing cells); and it is expected [2, 7] that, in the coming years, nickel consumption in this sector will demonstrate the highest growth rate (approx. 20% per year) of all applications for this metal. It is estimated that the production of various types of batteries consumed 36 thousand tons of nickel globally per year (including 65% in portable batteries). Intensive development of new types of batteries containing foam nickel, nickel hydroxide, or the La-Ni-Co alloy is also observed. This fact indicates that, in a short time, exhausted batteries (containing approx. 5–60% Ni) will constitute a major source of nickel recovery, especially that most of the nickel consumed in the country is imported. According to GUS data [7], the import of metallic nickel in recent years was nearly 2.6–2.9 thousand t/y (not including nickel compounds or scrap) at an annual consumption of 1.1–2.9 thousand tons (not including other forms of nickel and its compounds). For the last few years, only nickel sulfate (KGHM Polish Copper approx. 2,100 t/y) and small amounts of nickel alloys (up to 40 t/y) were produced in Poland.

Compared to both of the mentioned metals, up to 38% (approx. 27,000 t) of global cobalt demand is used for manufacturing various batteries (in form of powder, hydroxide or  $\text{LiCoO}_2$ ), which is the largest area of cobalt application (the second is the production of super alloys – 17%) [13]. In recent years, the largest increase in demand (40% per year) has been observed for lithium-ion batteries (containing 73%  $\text{LiCoO}_2$ ), first launched in 1991 by Sony. Their annual global sales reach several billion units. This is due to the rapid growth of the demand for mobile phones, laptops, and wireless devices, but also the development of the automotive industry (particularly, its most-advanced products like hybrid vehicles). In perspective, the majority of hybrid cars will be equipped with Li-ion batteries (currently, 95% of hybrid cars are powered by rechargeable Ni-MH batteries). Moreover, it is expected that, in 2020, the number of such vehicles will increase to almost 13 million [7]. Domestic demand for cobalt raw materials is completely met by import, since Poland does not produce this metal [1, 7].

The forecasted growth in global demand for lithium is mainly associated with the production of batteries (approx. 23%), glass fibers, special glass, and concrete with high resistance to thermal dilatation (ok. 31%) [14, 15]. It is expected that worldwide demand for lithium carbonate will maintain a growth rate of more than 50% and will reach 122,000 t in 2017. This is due to the development of the production of modern hybrid cars. Domestic demand for lithium raw materials (oxide, hydroxide, carbonate) is fully covered by import (in total approx. 220 t/y) [4]. They are used primarily in the glass,

ceramic, and electronic industries [14]. Lithium batteries are not recycled in Poland, but it seems that the production of even relatively small amounts of lithium carbonate could be consumed.

It should be noted that the report of the European Commission [16] confirms that cobalt is one of 14 “critical raw materials” for the European Union (also Sb, Be, Ga, Ge, etc.). Raw material is critical if the risk associated with a lack of its supply and its impact on the economy are higher compared with most raw materials. The report of the Organization for Economic Cooperation and Development [17] also includes nickel and lithium on the list of strategic raw materials.

### **3. Collection and recycling of batteries in Poland**

Due to the large amount and variety of batteries coming to the world market and the potential threat to the natural environment associated with the landfilling of the waste, problems of collection and the disposal of spent batteries has been the subject of legal regulations in the European Union. It introduced in 1991 the so-called “Battery Directive” (91/157/EEC), which was further amended (e.g., 93/86/EEC, 98/101/EC), and finally repealed by the currently applicable: Directive 2006/66/EC of 06.09.2006 (EU L 266) and a regulation No. 493/2012 dated 11.06.2012 (EU L 151/9). These regulations command all member countries to take actions to prevent threats from spent and expired electrochemical power sources, including organizing efficient collection systems, developing safe methods of waste disposal, and the obligatory utilization of all waste batteries and accumulators.

In Poland, the problem of waste management of batteries and accumulators is regulated by law. The Council of Ministers and Minister of the Environment have issued regulations since 2001 on the annual levels of recovery and recycling of packaging and post-use waste, including spent batteries and accumulators [3].

Several organizations on the recovery and recycling of batteries act in Poland; for example: Dol-Eko, Organizacja Odzysku SA, REBA Organizacja Odzysku SA, Organizacja Odzysku Eurobac SA, “Czyste Środowisko” Organizacja Odzysku SA. They associate the major domestic manufacturers and importers of batteries and accumulators. Educational campaigns and the collection of spent cells are organized not only as one-off nature actions [5], but also every entrepreneur placing the batteries market is required to achieve an appropriate level of collection. In addition, there is readily available information about the possibilities of the disposal of spent batteries or collection points (offices, schools, stores, etc.). In this case, the amount of waste collected depends largely on public awareness concerning the special handling of such materials (for example, Japanese studies [18] indicated that more than 70% of the batteries are not removed by the users from the

used or damaged electronic equipment like cameras, mobile phones, notebooks, etc.). Therefore, national legislation emphasizes education and information of the society and organizing a collection system of waste batteries and accumulators being a waste with a small size and a significant degree of distribution [19].

For several years in Poland, there has been an obligation to register the quantities of batteries and accumulators placed on the market and collected for recycling. Statistics led to the Chief Inspector of Environmental Protection GIOS [20]; but since 2014, reporting obligations have been taken by region marshals. Data provided by GIOS are not strictly detailed, because they are mainly focused on lead-acid accumulators, Ni-Cd, and zinc batteries placed on the market, while the recycling rate is calculated mainly for the first two groups (other types of batteries, including Ni-MH, lithium, silver, etc., are calculated cumulatively). On the basis of the values given for Ni-Cd batteries, only a rough estimation of the potential amount of nickel contained in the spent cells can be performed. In 2014, nearly 940 t of Ni-Cd (including approx. 606 t portable, about 70 t automotive, and approx. 262 t industrial) were placed on the domestic market [5], while two years earlier – approx. 716 t (approx. 489 t portable, approx. 1.2 t automotive, and approx. 226 t industrial) [6]. In 2014, Poland reached 85.5% of the recycling of waste Ni-Cd batteries and accumulators, while the required level was only 75%. However, it should be noted that Ni-Cd batteries are decreasing in the marketplace, and therefore such a waste stream will likely decrease about 2–3% per year [19].

The reached total collection level of waste batteries and accumulators was 33.06% in 2014 [5] (Fig. 1). However, when comparing the scale of introduced and collected spent batteries and the required levels of collection (45% in 2016 and the following years), a significant increase in the amount of recyclable waste material should be expected. According to reports and the GIOŚ registry [20], an increased number of entities placing the batteries on the market since 2010 (when the first report was published), while the number of entrepreneurs collecting/recycling waste batteries and accumulators remains practically at the same level. In the context of EU Commission Regulation No. 493/2012 (valid since 01.01.2014), all country members must fulfill requirements for minimal levels of recycling of used batteries and accumulators (e.g., 75% for Ni-Cd batteries). It should be noted that the recycling process does not include the steps of sorting and preparation for recycling (storage, handling, dismantling, etc.). In addition, the processing of different types of spent cells to acquire the relevant material fractions and the recycling of at least the extracted metals in the installations is required. Using the appropriate technology to ensure the achievement of at least the minimal levels of recycling (in a separate process or as part of a chain of events) is also requisite. This needs to determine the mass balance of the elements (chemical compounds) in the various technological processes [8].

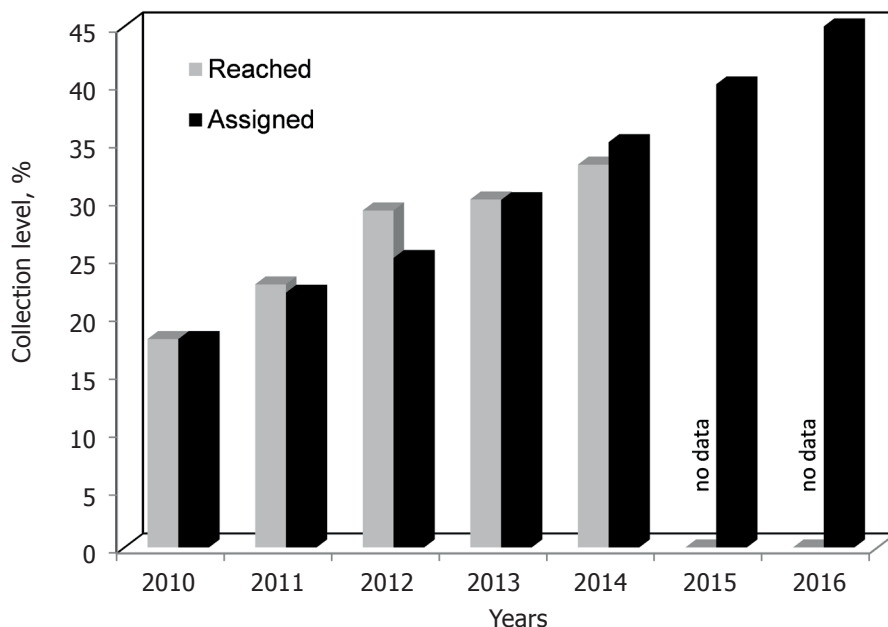


Fig. 1. Reached and assigned levels of spent battery and accumulator collection in Poland [19, 20]

Some of the countries have already implemented effective industrial recovery of metals from spent Ni-Cd, Ni-MH, or Li-ion batteries (Tab. 1), while there are no specialized plants focused on the recovery of nickel and cobalt in Poland. According to the GIOŚ register [20], 3153 operators are on the domestic battery and accumulator market, but only 24 entities collect waste batteries and accumulators (May 2016). The majority of them have R15 powers (i.e., “treatment of waste, in order to prepare them for recovery, including recycling”), but only 12 entities (Fig. 2) have R4 power (i.e., “recycling or reclamation of metals and metal compounds”) for the recovery of metals from batteries of different types; i.e., alkali, zinc, nickel (Ni-Cd, Ni-MH), lithium, and others, portable, automotive and/or industrial (excluding lead-acid cells). These are: Spółdzielnia Pracy ARGO-FILM (Warsaw; plant in Tarnów), MarCo Ltd sp. z o.o. (Katowice), Polska Grupa Recyklingu PROEKO sp. z o.o. (Legionowo-Lajski), MB Recycling sp. z o.o. Przedsiębiorstwo Gospodarki Odpadami sp. k. (Kielce), EKO HYBRES sp. z o.o. (Rogoźnica), RECUPYL POLSKA Sp. z o.o. (Gorzów Wielkopolski), EKO\_HARPOON Recykling sp. z o.o. (Cząstków Mazowiecki), HARPOON Sp. z o.o. (Cząstków Mazowiecki), 3MD Recycling sp. z o.o. (Legnica), BatEko sp. z o.o. (Wrocław), EKO-WYSZKÓW sp. z o.o. (Wyszaków), Biosystem SA (Kraków).



Fig. 2. Distribution and amount of domestic entities having R4 powers with respect to waste batteries and accumulators (excluding lead-acid ones) according to GIOS data (May 2016)

Two plants for processing waste Ni-Cd batteries and accumulators with a capacity of 2,000 t are in Poland [19]. Considering the mass of waste Ni-Cd batteries and accumulators recycled and the mass of batteries placed on the market in 2014, it is estimated that the capacity of the Ni-Cd cells recycling system is sufficient. However, according to the authors of the "National Waste Management Plan" [19], some recycling installations (especially the smaller ones) does not meet BAT requirements.

The recycling of Ni-Cd batteries is carried out in Huta "Oława." The plant operates a technological line for Ni-Cd and Fe-Cd cells that has been processing since 1995. After disassembly, nickel and cadmium electrodes, the electrolyte, and the housings are treated in separated processes: cadmium parts are converted pyrometallurgically into cadmium oxide (of a quality suitable for the production of cadmium-based pigments) and iron-nickel parts are used in the production of steels, while the electrolyte is used for the neutralization of acidic waste water [2, 7, 21].

Polish Recycling Group PROEKO recycles small- and large-sized Ni-Cd, Ni-MH batteries and accumulators, and Li-ion batteries, and the recovery of metals is conducted in France (the group cooperates with the French group S.N.A.M., a large producer of metallic cadmium), while collection of the waste is carried out in Poland [22].



French company Recupyl, together with Zakład Utylizacji Odpadów in Gorzów and AK NOVA sp. z o.o. (Poznań), built a plant for sorting and processing spent batteries with a capacity of 2,000 t/y. A technological line for recycling batteries was launched in Stanowice in 2010. The plant processes batteries from Sweden, Latvia, Lithuania, Germany, and Finland. RECUPYL POLAND sp. o.o. sorts and mechanically processes mainly zinc and alkaline batteries, while others type (e.g., Ni-Cd, Ni-MH, Li-ion, lithium) are sent to other plants (e.g., in Germany) [23].

Table 1. Exemplary recycling technologies of spent batteries and accumulators [24–28]

Company	Batteries/accumulators	Location
<b>Pyrometallurgical treatment</b>		
Dowa	All	Tokyo
Salesco Sytems	All	Phoenix
XStrata	All	Quebec, Nikkelverk, Sudbury
Toho Zinc	Ni-Cd, Ni-MH	Onahama
Japan Recycle Center	All	Osaka
SNAM	Ni-Cd, Ni-MH, Li	Saint-Quentin-Fallavier
NIREC	Ni based	Dietzenbach
Batrec AG	Li, Hg	Wimmis
AFE Group (Valdi)	All	Feurs, La Palais sur Veine
Citron	All	Zurich, Rogerville
Accurec	All	Mulheim
SAB NIFE	Ni based	Oskarshamn
Snam-Savam	Ni-Cd, Ni-MH, Li	Saint-Quentin-Fallavier
Umicore	All	Hoboken
Inmetco	Ni based	Ellwood City
<b>Hydrometallurgical treatment</b>		
OnTo Technology	Li	Bend
Euro Dieuze/SARP	All	Lorraine
IPGNA Ent. (Recupyl)	All	Grenoble
Korea Zinc	Ni-Cd	Onsan
Batenus	All	Schönebeck/Sachsen-Anhalt
<b>Physicochemical treatment</b>		
AEA Technology	Li	Sutherland
Toxco	Li, Ni based	Trawl, Baltimore

Since 2009, Ecoren DKE Plant in Polkowice has processed waste small-sized batteries and accumulators (annual throughput of 1,500 t) delivered mainly by the REBA organization. However, it was closed in April 2013 [29].

## 4. Summary

Requirements of the European Union for environmental protection and recycling levels state that “member countries shall encourage the development of new recycling and treatment technologies and promote research into environmentally friendly and cost-effective recycling methods for all types of batteries and accumulators” (Acts. Office EU L 266). However, in accordance with EU Regulation 493/2012, the recycling process does not include sorting and preparation for recycling (such as storage, handling, dismantling, and separation of fractions that are not part of the same batteries).

Polish analysis [2] confirms that the greatest challenges facing Polish society is “the adaptation of good practices related to the waste management of batteries,” which is an opportunity to “transform a recycling area into a modern and profitable branch of the national economy, providing a source of valuable raw materials”. What is more, the domestic economy of waste batteries and accumulators [19] assumed these as the most-important goals: the increase of public awareness and entrepreneurs about the correct way of treating the spent batteries and accumulators; achieving the appropriate levels of collection and recycling efficiencies in 2016 and in the following years.

Finally, the lack of more-detailed national data on the market and recycled cells other than Ni-Cd, zinc, and lead-acid should be given attention. Nevertheless, the market of such products is gradually being reduced in favour of advanced Ni-MH and lithium batteries. This generates the need to develop new technologies and to adapt recycling plants for the recovery of nickel, cobalt, lithium, and other valuable elements (lanthanides).

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