

Article

Life Cycle Impacts of Recycling of Black Mass Obtained from End-of-Life Zn-C and Alkaline Batteries Using Waelz Kiln

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Abstract: The utilization of end-of-life batteries (including Zn-C and alkaline batteries) is one of the areas that need to be perfected in order to provide environmental and human safety as well as to contribute to closing the material loop, as described in the EU Green Deal. The presented study shows the environmental impacts of the two selected pyrometallurgical technologies (processing of the black mass from waste Zn-C and alkaline batteries as an additive to an existing process of the recycling of steelmaking dust and treatment of the black mass as the primary waste material, both processes performed in a Waelz kiln). The presented LCA-based study of the recycling of end-of-life Zn-C and alkaline batteries focused on terrestrial ecotoxicity can be a useful tool in the process of the development of a circular economy in Europe, as it provides a multi-disciplinary overview of the most important environmental loads associated with the described recycling technologies. Therefore, the goal of the presented study was to compare the environmental performance (utilizing LCA) of two different metallurgical processes of black mass utilization, i.e., the conventional method utilizing black mass as a co-substrate and the newly developed method utilizing black mass as a primary substrate.



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1. Introduction

As described in the roadmap document published by Batteries Europe [1], the market for waste batteries is constantly growing. The most commonly used household batteries can be divided into two groups: primary (non-rechargeable, including Zn-C and alkaline batteries) and secondary (rechargeable, i.e., Ni-Cd, Ni-MH, Li-ion) [2]. In order to comply with the requirements for the implementation of the European Green Deal and provide resource independence from non-EU countries, it is necessary to develop modern and flexible solutions to processing different types of waste and recover as many secondary resources as possible. Alkaline and Zn-C batteries are the most popular types of non-rechargeable batteries commonly used in households to provide power to devices such as remote controls, alarm clocks, etc. [3]. Alkaline batteries represent as much as 72% of the total amount of batteries collected in the European Economic Area (EEA), while zinc-carbon batteries account for 7.7% of the total battery volume [4]. Landfilling of waste zinc-carbon or alkaline batteries can contribute to various environmental problems, as the heavy metals (i.e., Cd, Ni, Zn or Pb) can leach and cause contamination to the soil or water [5]. A scheme of Zn-C and the alkaline battery is presented in Figure 1. The cathode materials used in the batteries include MnO₂ and NH₄Cl for Zn-C and alkaline batteries, and LiCoO₂, LiNiO₂, LiFeO₂, Cr₃O₈ and manganese dioxide (MnO₂) are usually used for Li-ion batteries [6–8].

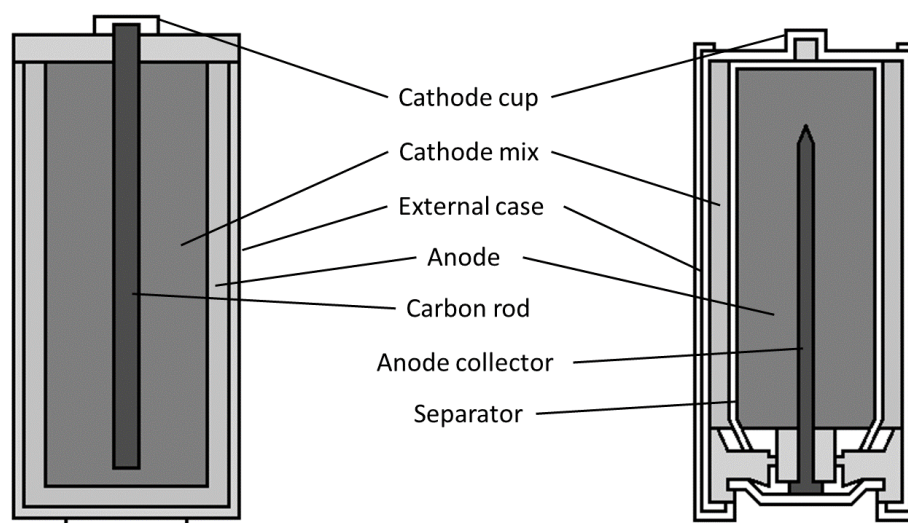


Figure 1. Scheme of Zn-C and alkaline battery [8].

The end-of-life batteries can be subjected to mechanical processing (crushing, magnetic separation and sieving) in order to prepare the materials for further processing: steel, plastics and black mass, which is an important source for the recovery of secondary zinc (Zn) and manganese (Mn) [9,10]. After mechanical separation, the black mass is sieved in order to remove any undesired coarse particles and can be recovered with the application of hydrometallurgical and/or pyrometallurgical processes. However, the effectiveness of the currently applied processes can be improved. Pyrometallurgical treatment of end-of-life Zn-C and alkaline batteries is usually performed at temperatures between 900–1500 °C and does not necessarily require the dismantling of the batteries. The processes are, however, energy-consuming and require the application of additional equipment in order to reduce the environmental loads [11]. Hydrometallurgical methods usually involve several steps, including pre-treatment, leaching and separation of metals. Those methods do not require the application of high temperatures, and no harmful substances are emitted into the air. The disadvantages are mainly the fact that the procedure requires multiple operations and the application of harsh chemicals and may lead to wastewater discharges [11]. A comprehensive approach to the recycling of end-of-life Zn-C and alkaline batteries was described by Tran et al. [12].

In recent years several attempts have been made in order to develop a new, more environmentally friendly method of black mass treatment. Some of them are based on solvent extraction utilizing different types of solvents and process parameters [9]. However, in most cases, a black mass is treated only as an addition to the other zinc-bearing materials. As its acquired volume gradually increases, modern metallurgical technologies should also be focused on the maximization of the amount of black mass used in the process.

A new waste-free technology based on the use of the Waelz kiln allows the black mass to be processed into two useful products: WOX (Waelz oxide) and manganese concentrate. This makes it possible to increase the recycling rate of batteries compared to the alternative technology since the manganese compounds are the product rather than being lost in the slag [13].

The benefits from this process come from the product as well as an environmental perspective. In order to confirm that and assess the real impact on the environment, a life cycle assessment (LCA) based analysis was performed. The LCA approach has proven to be a useful tool in environmental management as well as the decision-making process since it allows the detection of hotspots and promotes circularity [14–16].

2. Materials and Methods

LCA is a tool used to describe and quantify the environmental effects caused by products or processes in their whole life cycle, including mining, processing, manufacturing, transportation, distribution, utilization and recycling, expressed by the impact categories [17–20]. According to EN ISO 14,040, an LCA is “the compilation and evaluation of the inputs and outputs of a product system and its potential environmental impacts over its full life cycle” [18]. The environmental effects determined in the Life Cycle Impact Assessment (LCIA) phase of LCA can be presented at the midpoint or endpoint level. Midpoint characterization factors are, in general, associated with a lower level of uncertainty and can be used to present an impact in a single category (i.e., terrestrial ecotoxicity, land use or global warming), while the endpoint characterization factors, subject to larger levels of uncertainty, can be used to present the environmental impacts only in three damage categories: human health, ecosystems and resources [21]. A widely used LCIA method, ReCiPe 2016 [21], defines the toxicity midpoint categories as “environmental persistence (fate), accumulation in the human food chain (exposure), and toxicity (effect) of a chemical, (. . .) the cause-effect pathway, from emission to the environment, via fate and exposure, to affected species and disease incidences, leading finally to damage to ecosystems and human health”. The human toxicity impact category can be described as the increase in the risk of cancer (or non-cancer) disease incidence, and terrestrial, freshwater and marine ecotoxicity categories relate to the hazard-weighted increase in natural soils, fresh-or-marine water, expressed in kg of 1.4-DCB-eq (kilograms of 1.4-dichlorobenzene equivalent) emission to the respective medium [22].

The implementation of an LCA-based study of the recycling of end-of-life Zn-C and alkaline batteries focused on terrestrial ecotoxicity can be a useful tool in the process of the development of a circular economy in Europe, as it provides a multi-disciplinary overview of the most important environmental loads associated with the described recycling technologies. Therefore, the goal of the presented study was to compare the environmental performance (utilizing LCA) of two different metallurgical processes of black mass utilization, i.e., the conventional method utilizing black mass as a co-substrate and the newly developed method utilizing black mass as a primary substrate.

The calculations were performed using the ReCiPe 2016 Midpoint (H) and ReCiPe 2016 Endpoint (H) LCIA methods, SimaPro software (v. 9.2.0.2) and the ecoinvent database (v. 3.6). The ecoinvent database is considered to be one of the most reliable databases taking into account a large number of different processes [17,23]. The ReCiPe 2016 Midpoint (H) LCIA method evaluates a wide spectrum of impact categories related to toxicity and ecotoxicity, including terrestrial and marine ecotoxicity categories that rarely appear in other LCIA methods. The inclusion of these two ecotoxicity impact categories is crucial because of the significant emissions of metals (associated with the recycling of end-of-life batteries). Metals exhibit relatively high values of comparative toxicity potentials (characterization factors) and have a significant impact on LCA results [17].

2.1. Goal and Scope Definition

The goal of the presented study was to perform a gate-to-gate LCA in order to compare two pyrometallurgical technologies used to recycle black mass from waste Zn-C and alkaline batteries: a baseline Scenario 1 (steelmaking dust processed in a Waelz kiln with the addition of black mass as a co-substrate) and Scenario 2 represented with modern, waste-free technology (black mass processed in a Waelz kiln as the primary substrate). The goal was to determine whether the application of modern, waste-free technology will lead to the improvement of the recycling of end-of-life Zn-C and alkaline batteries as well as the reduction of any significant environmental loads, with a special focus on toxicity and ecotoxicity. It is expected that the application of waste-free technology described in scenario 2 will result in significant reductions in impact categories associated mostly with eco- and human toxicity since no slag is stored in landfill and leaching of heavy metals is avoided. In both cases, the functional unit was the processing of 1Mg of black mass. The recycler

is not capable of influencing the initial life-cycle steps of the Zn-C and alkaline batteries, such as the manufacturing or use phase. The products are also designed to be sold to a third-party company. The system boundary for both scenarios is presented in Figure 2.

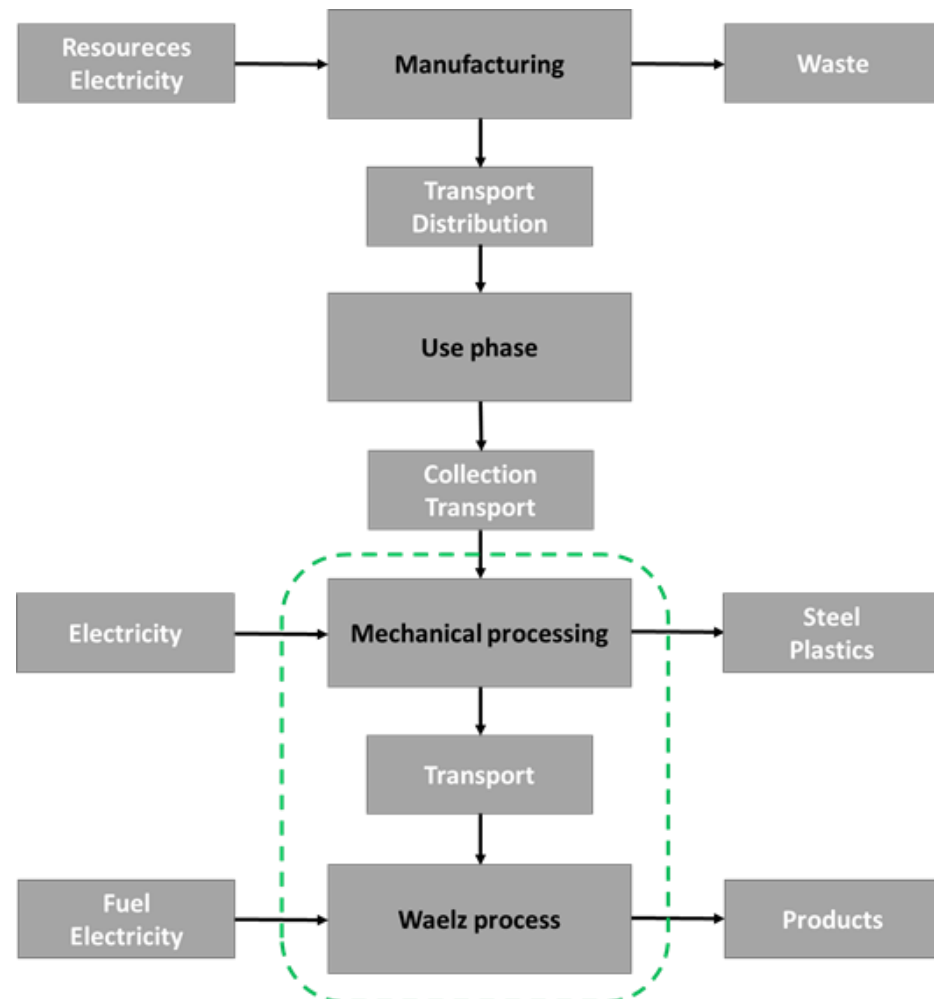


Figure 2. System boundary.

Due to the lack of impact of packaging on the outcome of the black mass recycling process (as it is identical in both scenarios), it was skipped for the purpose of the analysis. However, it should be noted that this analysis focuses on the recycling method of the end-of-life product, whereas if the environmental impact of the cells themselves is considered, the production and disposal of packaging would have to be included.

2.1.1. Scenario 1—Black Mass as a Co-Substrate in Steelmaking Dust Processing in a Waelz Kiln

The black mass can be directed to the electric arc furnace (EAF) dust treatment process in the Waelz furnace. Dust from an EAF dust collector containing approximately 30% ZnO and 35% FeO is treated in this process. The addition of black mass to the process is limited to 20%. The main disadvantages of diverting black mass to the Waelz process include the recovery of Zn only and the loss of Mn in the slag due to the low Mn concentration [22]. In the Waelz process, the zinc-bearing input material is processed at approximately 1200 °C with the addition of a reducer. This yields Waelz oxide (WOX), containing mainly ZnO, and slag rich in Fe oxides. WOX is used for the production of zinc. On the other hand, slag containing also Mn compounds besides Fe (coming from black mass) can be used as road aggregate [24].

Only the black mass components that are transferred to WOX (Zn compounds) will be included in the recycling efficiency. The components that are transferred to the slag (including manganese oxides) are not recovered. Due to the lack of experimental data, the information regarding the mass flows of the process described in scenario 1, as well as the composition of EAF dust, oxide products, produced slag and off-gases, are based on literature [25,26].

2.1.2. Scenario 2—Waste-Free Technology of Black Mass Processing in Waelz Kiln

The waste-free technology of black mass processing in Waelz kiln was described in a Polish patent [13]. The goal of the process is to produce Mn concentrate in the form of sinter (slag) and Zn concentrations in the form of dust. Its main advantage is the processing of the non-magnetic waste fraction, generated from the mechanical processing of zinc-carbon and alkaline batteries, in a single technological operation of reductive roasting into two valuable products: zinc concentrate in the form of dust collected from the gas-dedusting process and manganese concentrate, containing mainly MnO, obtained as a furnace product in the form of slag, with zinc and manganese yields over 85%. The inputs in the process are a non-magnetic fraction generated during the mechanical processing of zinc-carbon and alkaline batteries and the addition of a reducer in the amount of 10–30% of the weight of the metal-bearing input.

2.2. Life Cycle Inventory Analysis (LCI)

One of the most important factors that may affect an LCA study is the lack of data and its uncertainty. Sometimes the primary data can not be provided, i.e., due to confidentiality reasons. Sometimes the data acquired from databases are not relevant, which complicates the calculations for a comparative study. It is crucial to use as much primary data as possible when performing an LCA analysis. However, in the majority of cases, the inventoried data only partially describe all of the emitted substances, as their full identification is often impossible or technically demanding. These factors may have a significant influence on the obtained LCA results, especially when the comparison is made between products/processes utilizing internally measured data and products/processes utilizing data from external databases [27,28]. In the presented study, however, similar amounts of emitted data were inventoried for both scenarios during real-life operations of the devices.

Based on the primary data provided by the recycling company as well as the literature [26], it was assumed that mechanical treatment of spent zinc cells to produce black mass requires 160 kWh of electricity per 1 Mg of black mass. In addition, 2.083 Mg of cells is required to generate 1Mg of black mass.

The processing of 1 Mg of black mass requires the consumption of 20 m³ of natural gas and 200 kg of coke with a calorific value of 30 MJ/kg. These data are based on 70 years of experience gained by the researchers working in Łukasiewicz Research Network-Institute of Non-Ferrous Metals and presented in the internal reports. The distance between the mechanical processing site and the metallurgical plant was assumed to be 860 km (this is the average of three potential sites in Germany (Relux, Bad Oeynhausen; Redux, Bremenhausen; and Redux, Offenbach) and Kędzierzyn-Koźle, Poland-the potential location of the plant). The value of electricity consumption (175 kWh) during metallurgical treatment of 1 Mg of black is based on literature [25].

The life-cycle inventory analysis for Scenario 1 (black mass as a co-substrate in steel-making dust processing in Waelz kiln) and Scenario 2 (waste-free technology of black mass processing in Waelz kiln) is presented in Table 1. Other data necessary to perform the calculations are presented in the supplementary material (Tables S1–S3).

Table 1. Life-cycle inventory: Scenario 1—black mass as a co-substrate in steelmaking dust processing in Waelz kiln, and Scenario 2—waste-free technology of black mass processing in Waelz kiln.

LCI	Per 1 Mg of the Processed Black Mass:			
	Input	Unit	Scenario 1	Scenario 2
Emissions to air	Oxygen, O ₂	kg	168.1	2.1
	Carbon dioxide, CO ₂	kg	605.1	13.67
	Carbon monoxide, CO	kg	33.6	3.4
	Hydrogen, H ₂	kg	-	2.4
	Nitrogen, N ₂	kg	2386.9	106.5
	Steam	kg	134.5	-
Material consumption	Natural gas	m _n ³ /Mg	-	20
	Coke	MJ	23,969.4	6000
	Black mass	kg	1000	1000
	Water	kg	802.9	-
	Steelmaking dusts	kg	4662.5	-
	Limestone	kg	543.6	-
	Oxygen, O ₂	kg	1509.6	-
	Electricity	kWh	175	175
Products	Zn concentrate	kg	-	393
	Mn concentrate	kg	-	479
	Oxide material	kg	2118.9	-
	Waste slag	kg	3836.9	-

3. Results and Discussion

3.1. Scenario 1

The characterization results for Scenario 1 for all 18 impact categories analyzed using the ReCiPe 2016 Midpoint (H) method are presented in Figure 2. For the majority of the impact categories, the highest contribution (21–96%) in the total impact score can be associated with the production and processing of black mass. This may be explained by the inclusion of various technological processes such as mechanical processing (primarily due to the high consumption of electricity produced from fossil fuels), mining and production of metals (especially zinc oxide, copper, brass, ferronickel, manganese dioxide or ferrochromium), and, in the case of the ionizing radiation impact category, the operation of a nuclear power plant. The lowest contribution (i.e., 21–69%) of black mass production and processing was particularly relevant for toxicity and ecotoxicity impact categories (with the exception of terrestrial ecotoxicity with 82% of contribution), where significant contribution (28–77%) was observed for landfilling of zinc slag. This is a result of the significant emission of metals to soil and water during landfilling of slag. Metals, in general, exhibit the highest values of characterization factors (CF) across many substances considered in all impact categories related to toxicity and ecotoxicity [17]. Also, in many cases, the highest contribution in total impact scores can be associated with the application of coke (0.1–22%). Coke had a significant contribution in fossil resource scarcity (22%), fine particulate matter formation (18%), stratospheric ozone depletion (17%) and ozone formation (13–15%) impact categories. This shows the importance of the development of modern non-coke-based metallurgical technologies in order to decarbonize the European industry and meet the European Green Deal requirements [22,25,26,29–31]. The diagrams showing the calculated environmental impacts for all the processes (including the influence of the black mass itself) are presented in Figures S1 and S2 in the Supplementary Material. The influence of all the remaining processes (black mass excluded) is presented in Figure 3.

Scenario 1, ReCiPe 2016 (H)

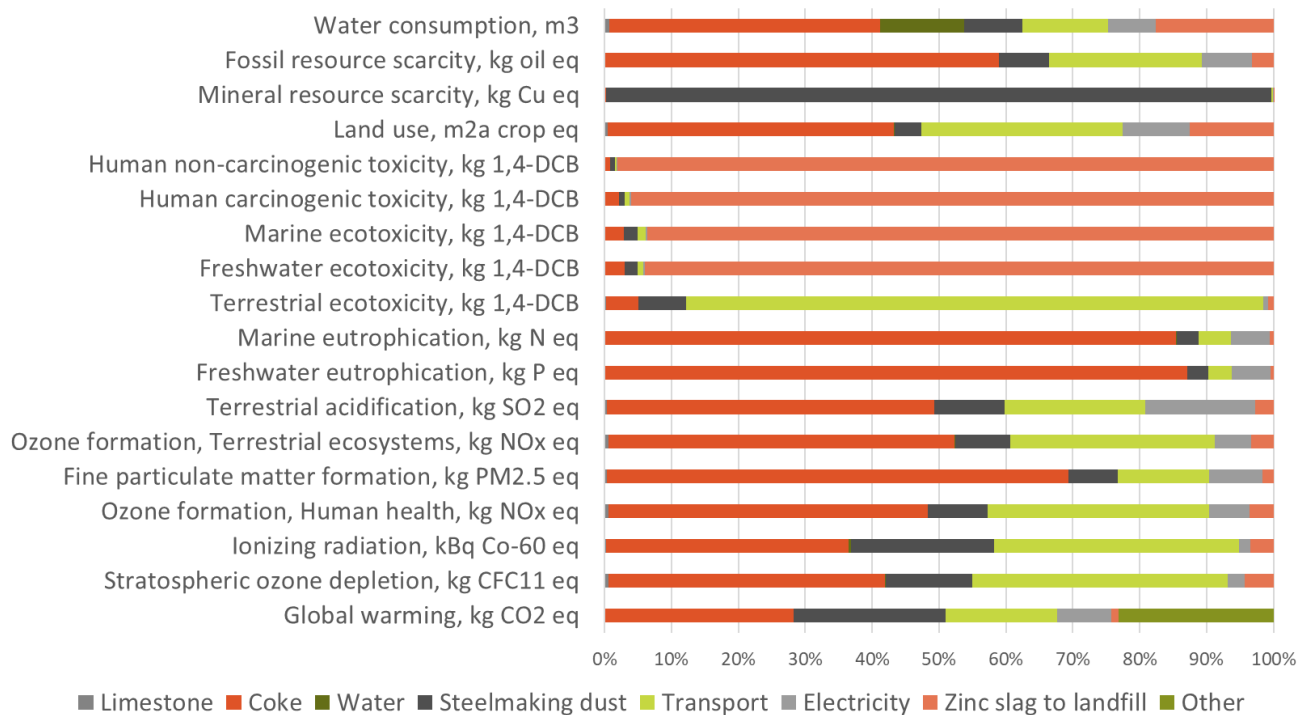


Figure 3. The contribution of the individual process in total impact scores was obtained for 18 different impact categories for scenario 1 (black mass excluded).

3.2. Scenario 2

The characterization results for Scenario 2 for all 18 impact categories that were analyzed with the ReCiPe 2016 Midpoint (H) method (influence of black mass excluded) are presented in Figure 4. Similarly, as observed in Scenario 1, the highest contribution in total impact scores across all impact categories can be assigned to the black mass. In this case, however, the impact of other combined processes, such as the application of coke, electricity and transport, is significantly lower, as the contribution of black mass ranged from 84 to 99%. This may be explained by lower consumption of coke and the avoidance of landfilling of zinc slag.

3.3. Comparison

The comparison between Scenario 1 and Scenario 2 in terms of their total impact scores for each impact category is presented in Figure 5. It can be observed that for each impact category, the environmental impact of Scenario 2 is lower, which is especially evident for human non-carcinogenic and cancerogenic toxicity impact categories, but also mineral resource scarcity. Similar results were obtained using the ReCiPe 2016 Endpoint (H) method considering damage assessment, where Scenario 1 exhibited a higher impact on human health, ecosystem and resources (Figure 6).

Scenario 2, ReCiPe 2016 (H)

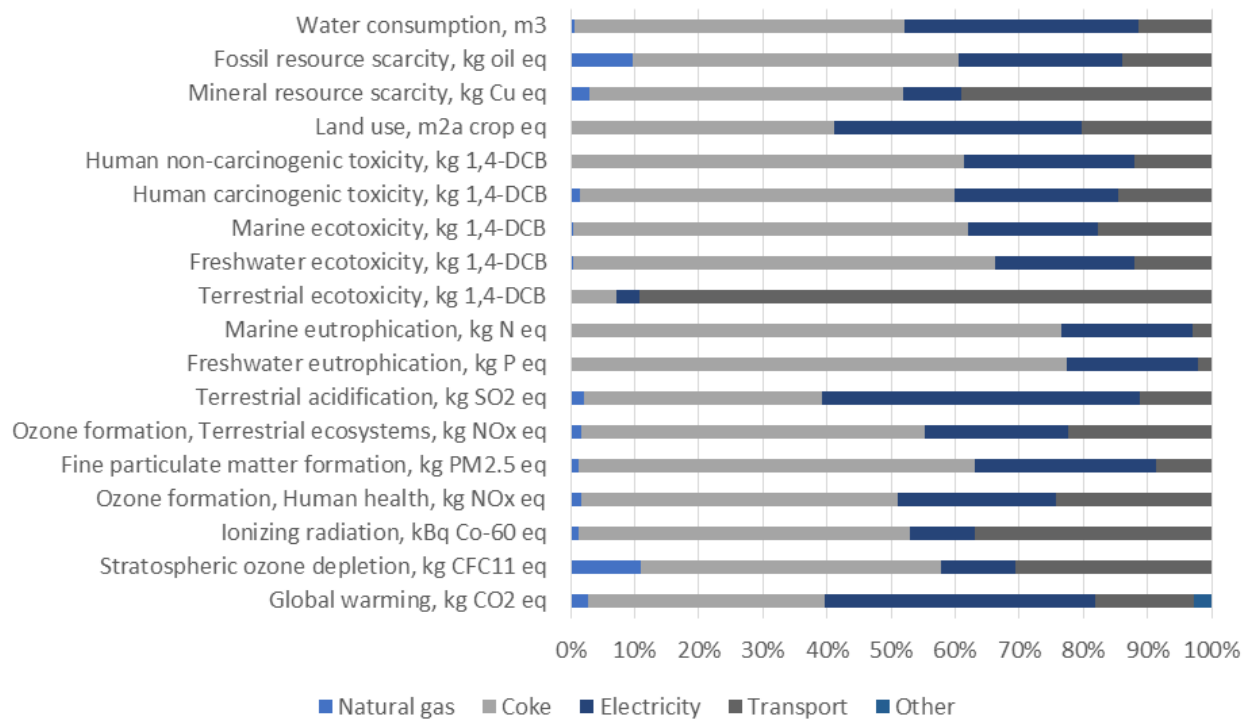


Figure 4. The contribution of various combined processes in total impact scores was obtained for 18 different impact categories for scenario 2 (black mass excluded).

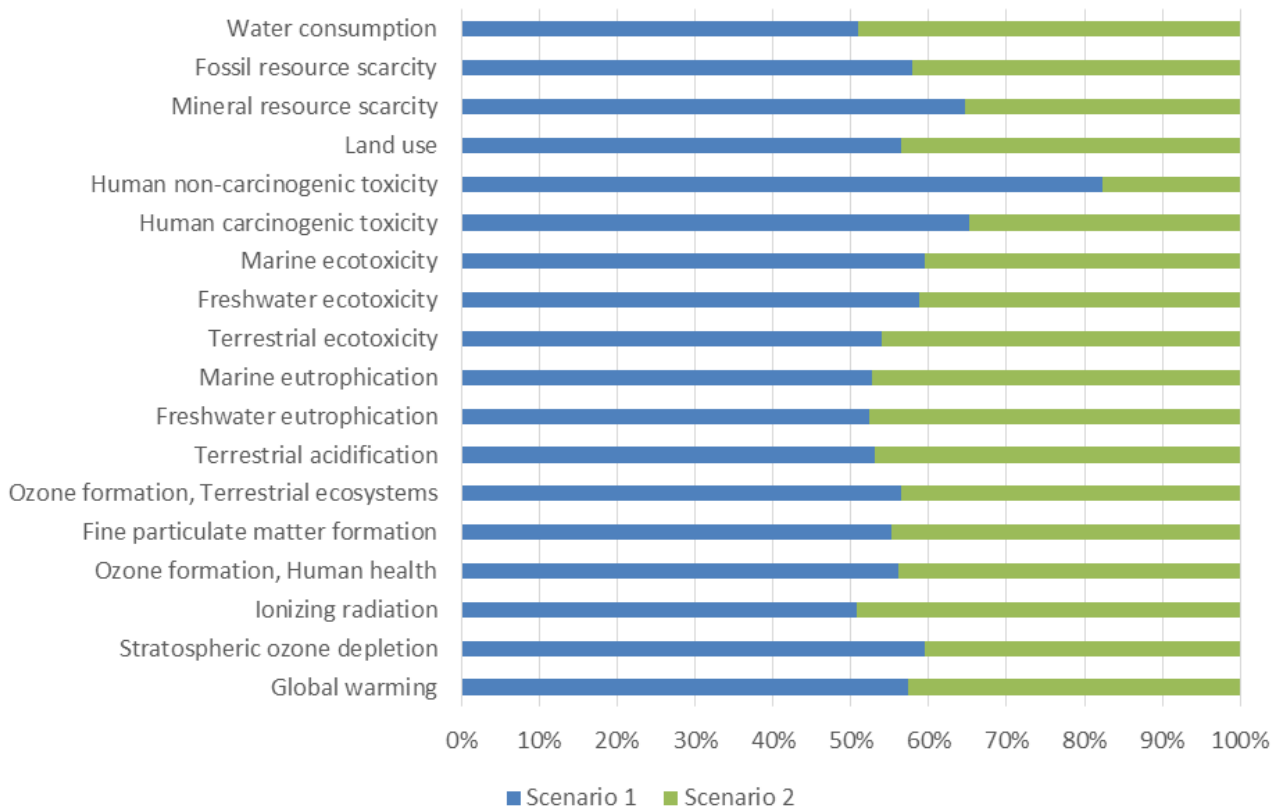


Figure 5. Comparison of the environmental performance of both studied scenarios in terms of their dominance in all of the 18 impact categories available in the ReCiPe 2016 Midpoint (H) method.

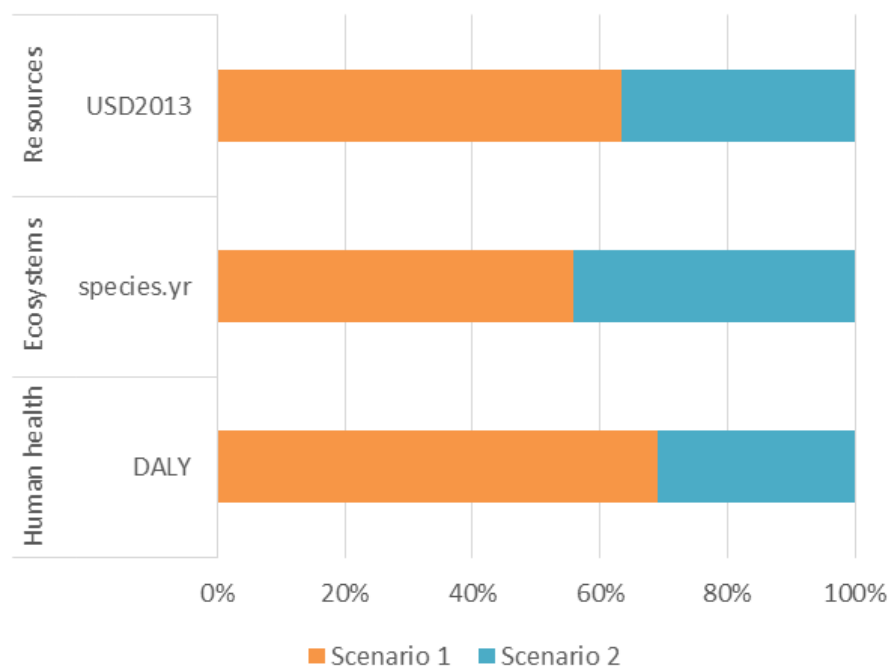


Figure 6. Comparison of the environmental performance of both studied scenarios in terms of their dominance in damage assessment categories available in the ReCiPe 2016 Endpoint (H) method.

After normalization, it is clear that the toxicity and ecotoxicity impact categories have the greatest influence on the results of performed LCA analysis with the particular dominance of the human carcinogenic toxicity impact category (Figure 7). In this impact category, regardless of the analyzed scenario, the single dominant substance is Cr (VI) emitted to water, which is responsible for 93–96% of the obtained values of the total impact score. On the other hand, in the human non-carcinogenic impact category, the dominant substances, regardless of the considered scenario, are Zn (emitted to water) and As (emitted to water), which combined are responsible for 85% (Scenario 2)–96% (Scenario 1) of the obtained values of total impact score. Similar results regarding the dominant contribution of metals in total impact scores were obtained for ecotoxicity impact categories – freshwater ecotoxicity was dominated by the emissions of Cu and Zn to water (89–92% depending on the scenario), marine ecotoxicity was dominated by the emissions of Cu and Zn to water (88–90% depending on the scenario), and terrestrial ecotoxicity was dominated by the emissions of Cu and Zn to air (84–87% depending on the scenario). Complete results are presented in the Supplementary material (Figures S2–S7). These results clearly show how important the emissions of metals are in all impact categories dedicated to toxicity and ecotoxicity. Metals usually exhibit one of the highest characterization factors (also called comparative toxicity potentials) among thousands of substances included in various LCIA methods (including ReCiPe 2016) [19]. Therefore, the influence of metals on the final results of LCA is especially important in all products/technologies/processes that can possibly generate significant emissions of metals to air, water and soil, such as metallurgical or agricultural processes [17,32]. Additionally, as the bioavailability of metals emitted to soils is limited, e.g., due to their interactions with the components of the soil (organic matter, etc.), it is clear why the terrestrial ecotoxicity impact category exhibited lower influence on the final LCA results as compared to other toxicity and ecotoxicity categories [33].

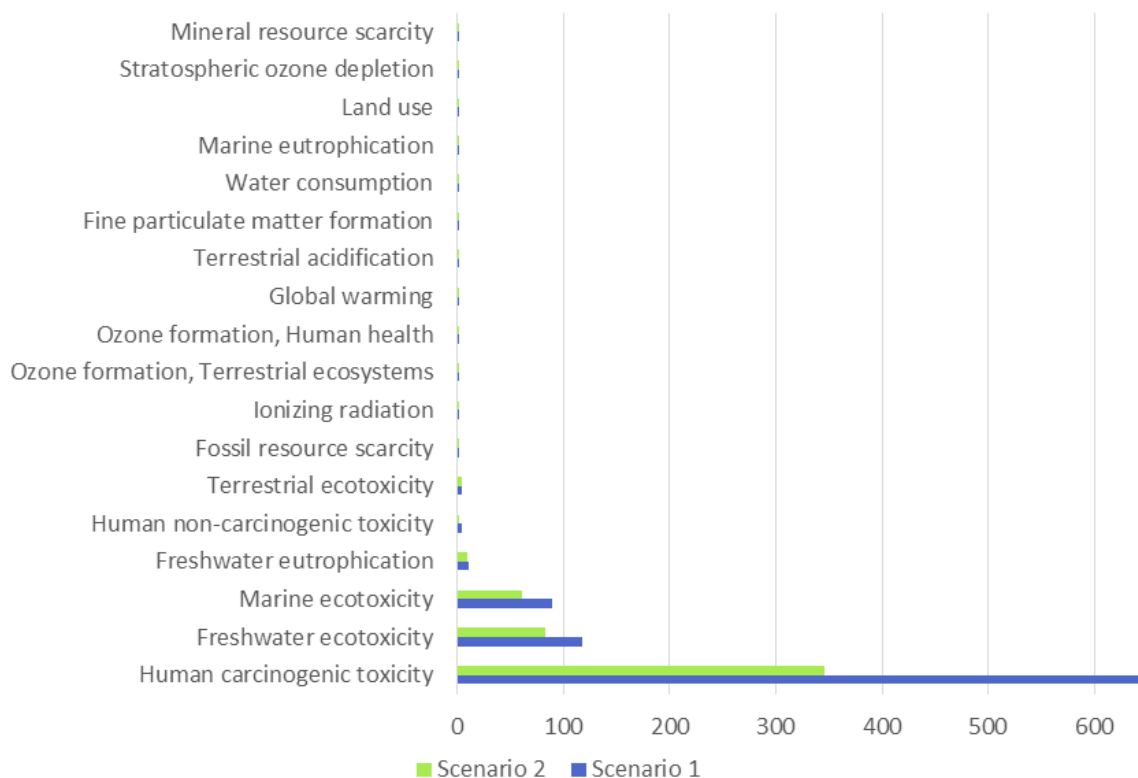


Figure 7. The comparison between scenario 1 and scenario 2 after normalization of the obtained results.

4. Summary

The growing market for end-of-life Zn-C and alkaline batteries forces the EU member countries to develop and implement modern recycling technologies in order to recover valuable components of this waste as well as reduce the negative, hazardous impact of waste batteries on the environment as well as human health.

The presented study was performed in order to evaluate the weak points of two similar recycling technologies that utilize the Waelz kiln to process waste batteries. The first technology (Scenario 1) allows the processing of black mass from waste batteries as an additive to steelmaking dust recycling. The second presented technology (Scenario 2) is a waste-free technology, where the black mass is processed into Zn and Mn concentrates. The obtained results clearly indicate that the waste-free scenario, which simultaneously exhibited lower consumption of coke, has a lower environmental impact regardless of the analyzed impact category. Additionally, the emission of metals during landfilling of zinc slag was one of the main factors differentiating both scenarios. At the same time, in the most important scenario in terms of the obtained results, it seems that all of the future battery-waste utilization methods should focus on the reduction of the metal content in the residual waste materials.

Technologies of battery processing are developing all the time, but it is crucial that the recycling process should also be beneficial for the environment. The recycling process can be perfect with the products in terms of quality and quantity, but the most important indicator should give the picture that this technology is friendly to the environment. After performing of both LCA and economic assessment and achieving satisfying results from both perspectives, the implementation of technology could appear in real conditions. Battery recycling technologies are very demanding in the current stage. Therefore, further searching for zero-waste solutions seems to be an important factor influencing the development of this market in the future.

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/en16010049/s1>, Table S1: Black mass composition; Table S2: Waelz oxide (WOX) composition [29]; Table S3: Summary of the inputs and outputs for the mechanical processing; Figure S1: The contribution of the individual process to total impact scores as obtained for 18 different impact categories for Scenario 1; Figure S2: The contribution of various combined processes in total impact scores as obtained for 18 different impact categories for Scenario 2; Figure S3: Terrestrial ecotoxicity; Figure S4: Freshwater ecotoxicity; Figure S5: Marine ecotoxicity; Figure S6: Human carcinogenic toxicity; Figure S7: Human non-carcinogenic toxicity.

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Conflicts of Interest: The authors declare no conflict of interest.

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