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Procedia CIRP 48 (2016) 188 – 193

www.elsevier.com/locate/procedia

23rd CIRP Conference on Life Cycle Engineering

The Environmental Impacts of Recycling Portable Lithium-Ion Batteries

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Abstract

Waste from electronic and electrical equipment (WEEE) is one of the fastest growing waste streams, with its volume expected to increase by a third from 2013 to 2017. Lithium-ion batteries are the most common battery type used in portable electronic devices and their use is expected to double from 2013-14 to 2019-20. The recycling of lithium-ion batteries reduces energy consumption, reduces greenhouse gas emissions, and results in considerable natural resource savings when compared to landfill. However, it is unclear which recycling processes have the least impact on the environment. This paper will investigate the different processes that are currently used for recycling portable lithium-ion batteries, such as hydrometallurgy, pyrometallurgy, and combinations of processes. Surveys are carried out to understand the materials recovered from each process, and are obtained from several recycling companies around the world. A comparative life cycle assessment will be performed for two different recycling processes (hydrometallurgy and pyrometallurgy), in order to understand the associated environmental impacts. This study shows that the largest contributors to the environmental impacts are electricity generation, incineration of plastics, and landfilling of residue. In terms of environmental effects, it is suggested that the most beneficial processes are those that utilise low temperatures, and are capable of recovering plastic.

Peer-review under responsibility of the scientific committee of the 23rd CIRP Conference on Life Cycle Engineering

Keywords: Life cycle assessment; lithium-ion batteries; recycling

1. Introduction

Waste from electronic and electrical equipment (WEEE) is one of the fastest growing waste streams, with its volume expected to increase by a third from 2013 to 2017 [1]. An increasing amount of waste requires more land area for disposal, and adds to the amount of harmful chemicals that eventually re-enter the environment [2]. The increasing use of portable electronic devices is also the increasing disposal of portable batteries that consist of various toxic substances. Lithium-ion batteries are the most common battery type used in portable electronic devices and their use is expected to double from 2013-14 to 2019-20 [3]. These batteries contain lower levels of toxic materials than other battery types [4], and in some countries, are considered suitable for disposal to landfill.

It has been shown that recycling batteries is beneficial to the environment. Recycling lithium-ion batteries in particular reduces energy consumption [5], reduces greenhouse gas emissions, and results in 51.3% natural resource savings when compared to landfill [6]. The majority of benefits occur as a result of avoiding virgin materials production [7]. However it is not possible to recycle lithium-ion batteries without causing any environmental impacts [8].

Currently in Australia, no recycling of lithium-ion batteries is performed, and 98.3% of lithium-ion portable batteries end up in landfill [3]. The Australian Battery Recycling Initiative (ABRI), a not-for-profit association, is working to achieve increased recovery of all battery types and responsible environmental management in the battery recovery chain. MobileMuster is the Australian mobile phone industry's official product stewardship program to collect end-of-life mobile phones (including batteries) for recycling. These organisations must make decisions regarding where these batteries are recycled. Factors influencing these decisions may be costs, recycling efficiencies and environmental effects.

The aim of this project is to independently investigate the different processes that are currently used for recycling lithium-ion batteries, and to compare these processes in terms of recovered materials, costs, efficiency and environmental effects; as well as to compare the effects between recycling

Peer-review under responsibility of the scientific committee of the 23rd CIRP Conference on Life Cycle Engineering
doi:10.1016/j.procir.2016.03.100

and landfill. This information can be used by organisations such as the ABRI and MobileMuster to ensure the most appropriate disposal options are selected.

1.1. Background

Lithium-ion battery composition varies with size, application and cathode material. Several composition analyses were collated to form a composition breakdown of a typical lithium-ion portable battery, shown in Figure 1. The base composition was taken from secondary sources [9][10], and was adjusted based on correspondence with battery recycling companies. It was assumed that the battery consists of a lithium cobalt oxide cathode and an iron-nickel alloy casing.

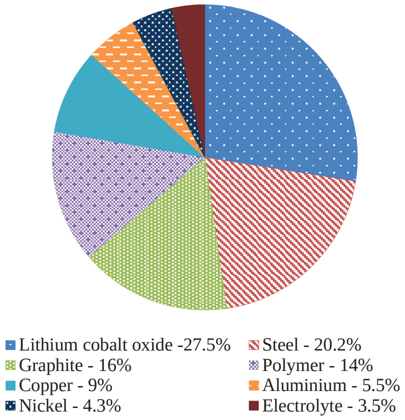

Figure 1. Lithium-ion battery composition

This composition was used to estimate the value for one tonne of waste batteries. This is shown in Table 1.

Table 1. Estimated value of materials

Material	Price ($AUD/tonne) [11]–[13]	$AUD available/tonne batteries
Nickel	18684.00	803.40
Aluminium	2464.00	135.55
Copper	8168.00	735.10
Steel	567.00	114.60
Lithium cobalt oxide	36370	10001.75

Techniques for recycling portable batteries can be broken into three general categories: mechanical, pyrometallurgical and hydrometallurgical processes. Often, a combination of these techniques is used in succession for the recovery of different materials.

Mechanical processes have two purposes. The first is to dismantle the battery and liberate components. These processes may include crushing and shredding [14]. They are also used to separate crushed components, by sorting materials according to their physical properties [15]. These processes may include magnetic separation, air ballistic separation and sieving.

Hydrometallurgical processes recover metals using acids or bases to leach metals into a solution, which is subsequently purified to extract the materials [15]. These processes are preceded by a mechanical process such as crushing or shredding to liberate the materials [14]. Hence where processes are referred to as hydrometallurgical, this includes the mechanical pre-treatment.

Pyrometallurgical processes use high temperatures to recover materials. These processes may include pyrolysis, smelting, distillation and refining. Lithium and organic compounds such as paper and plastic are not recoverable using pyrometallurgical processes alone [14].

Similar research has been undertaken to compare these recycling processes. Sangwan and Jindal [16] provided a decision model for evaluating recycling alternatives for lithium-ion batteries against a range of criteria. However, their work was non-quantitative and relied on expert opinion to rank processes against the criteria. Little investigation was done into the environmental impacts, and transport associated with recycling was not considered. This study focuses on environmental impacts, and undertakes life cycle assessment, which quantifies the effects of recycling lithium-ion batteries.

2. Methodology

A list of eleven companies currently recycling lithium-ion batteries was compiled based on contacts of the ABRI and MobileMuster, as well as further independent research. A survey was sent to these companies requesting information regarding the processes used and the materials recovered. Of the eleven, six responded with sufficient data, and were also able to provide further information on the material inputs and outputs of the recycling processes. Data for two additional companies was available through secondary sources, giving a total of eight companies included in the analysis.

For the environmental impact component of the analysis, life cycle assessment (LCA) was used. The product was defined as portable lithium-ion batteries, and the goal was to provide a comparison between different processes for recycling these batteries. The functional unit was 1 tonne of batteries, and all values were determined in terms of this unit.

The scope of the analysis included the end-of-life phase of the product life cycle only, and collection was excluded from the analysis. The impact categories chosen were: global warming potential over a 100 year time period (GWP 100), human toxicity potential (HTP) and terrestrial ecotoxicity potential (TETP). GWP 100, expressed in kilograms of carbon dioxide equivalent (kg CO2-eq) was chosen due to the current importance of assessing the effects of current processes on global warming. HTP and TETP, both expressed in kilograms of dichlorobenzene equivalent (kg DCB-eq) were chosen due to the end-of-life focus of the analysis. Currently, most lithium-ion batteries are sent to landfill, where they can leach materials to the surrounding environment.

GaBi LCA Software was used for the assessment, and all characterisation was performed using the CML 2001-April 2013 database. Normalisation of the results was also performed to compare the results to a reference value: the impact of one person in one year. For this normalisation, the 'World, Year 2000' factors were used. This gives the results in terms of person equivalents (PE), or the impact potential per person per year, without specific reference to one region.

Table 2. Survey results: Processes and recovered materials

Company	Process	Location	Cu	Al	Ni	Li	Co	Mn	Steel/iron	Plastic	C	Fl
P1	Pyrometallurgical	Europe	Y	Y	Y	N	Y	Y	Y	N(i)	Y(a)	N
P2	Pyrometallurgical	Europe	Y	Y	N	N	Y	N	N	N	N	N
M1	Mechanical	Europe	Y	Y	Y	N	Y	Y	Y	Y(u)	N	N
C1	Combination	Europe	Y	N	Y	N(c)	Y	N	Y	N	N	Y
C2	Combination	Europe	Y	Y	Y	N	Y	N	Y	Y(r)	N	N
H1	Hydrometallurgical	Asia	Y	Y	N	N	Y	N	Y	Y(r)	N	N
H2	Hydrometallurgical	America	Y	Y	Y	Y	Y	Y	Y	N(l)	N	N
H3	Hydrometallurgical	Asia	Y	Y	Y	N	Y	Y	Y	N(l)	N	N

(r) = further recycled, (l) = landfill, (c) = addition to concrete, (a) = reused as reduction agent, (i) = incinerated with energy recovery, (u) = unspecified

3. Results

3.1. Recovered materials

Through a combination of survey results and secondary sources, data concerning the process used and the materials recovered was obtained for eight different recycling companies globally. See Table 2 for the results. A 'Combination' of processes refers to companies that use both hydrometallurgical and pyrometallurgical processes to recover materials from lithium-ion batteries.

The results show that all companies included in the analysis recover copper and cobalt. This result was expected due to the high value of these materials. Steel, nickel and aluminium were also found to be commonly recovered. Although steel represents the lowest value component of the battery, it is one of the simplest to extract, since it can be separated magnetically if mechanical processes are utilised at the first stage of the recycling process. Similarly, aluminium has lower value, but is still recovered. This is likely due to the demand for recycled aluminium, considering the high cost and energy requirements of producing aluminium from raw materials.

Most companies claimed to recover plastic, with those that did not claim recovery utilising pyrometallurgical processes at first stage, which burns the organic material. The remaining companies either recycled, landfilled, or incinerated for energy recovery. Despite survey results, if plastics were incinerated or landfilled they were not considered recovered. The remaining materials, lithium, manganese, carbon and fluorine were found to be not commonly recovered. A summary of the survey results is shown in Table 3.

Table 3. Survey results: Distribution of processes used

Process	Number of companies	Average number of recovered materials
Hydrometallurgical	3	6
Pyrometallurgical	2	5
Mechanical	1	7
Combination	2	6

Overall, from the survey results it can be seen that purely pyrometallurgical processes recover the lowest number of materials. These processes are the most flexible in terms of input, but the extracted materials cannot easily be adjusted. On the other hand, hydrometallurgical processes are more specific to the battery type and hence are capable of recovering a larger number of materials [17].

Using a combination of hydrometallurgical and pyrometallurgical processes showed that on average, the same number of materials can be recovered when compared to purely hydrometallurgical processes. Pyrometallurgical process is often used as a pre-treatment before the leaching process to remove impurities such as organic matter, thus no additional materials are recovered for this additional step [18].

Mechanical processes were shown to recover the highest number of materials. The survey results indicated that materials extracted from mechanical processes are often sent to specialised recycling facilities for refinement. No survey response was received for Company M1 and hence the results are obtained through secondary sources only. The available information did not indicate whether the recovered materials are from Company M1's process alone, or if they refer to materials recovered in stages performed by other companies further down the line. Hence, it is unclear whether recovery can be performed using mechanical processes only.

3.2. Costs

The recycling companies were asked through the survey to indicate whether they charged a fee to collectors for recycling, or if they paid collectors for spent batteries. The responses are shown in Table 4.

Table 4. Survey results: Payment types

Company	Process	Location	Payment type
P1	Pyrometallurgical	Europe	Pays for high-cobalt batteries, charges for low-cobalt batteries
P2	Pyrometallurgical	Europe	Charges for batteries
C1	Combination	Europe	Pays for lithium-ion vehicle batteries per piece
C2	Combination	Europe	Charges for rechargeable lithium batteries, pays cobalt valorization if content >6%
H1	Hydrometallurgical	Asia	Pays for batteries containing cobalt

Table 5. Environmental impacts due to transport

Location	Distance (by sea)	GWP 100		HTP		TETP	
Units	*km*	*kg CO_2-eq*	*PE*	*kg DCB-eq*	*PE*	*kg DCB-eq*	*PE*
Europe (Rotterdam)	21428	306	7.3e-12	14.1	5.5e-12	0.0446	4.1e-14
North America (Houston)	17112	245	5.9e-12	11.2	4.4e-12	0.0356	3.3e-14
Asia (Singapore)	7914	113	2.7e-12	5.2	2.0e-12	0.0165	1.5e-14
Australia (Sydney)	0	0	0	0	0	0	0

The results show a strong relationship between recycling and value of materials, with batteries containing cobalt generally being bought by recyclers. Most companies indicated that they buy waste batteries for processing. However, in Europe, where collection and recycling efficiency targets are enforced, recycling companies may be willing to accept batteries that do not contain cobalt in order to meet these targets. The result is that they must charge a fee for this service. In other locations, where there are no targets, recycling is purely price driven, so companies are more likely to only accept valuable batteries.

3.3. Efficiency

The surveys sent to recyclers also requested the recycling efficiency by weight associated with the processes used. Most companies were unable to provide this due to privacy reasons. Using the assumed composition and materials recovered, a maximum possible recycling efficiency was calculated for each company. These efficiencies were optimistic, assuming that any recovered plastic was further recycled (unless otherwise specified), and assuming each material was completely recovered. Carbon was not included in these calculations, due to the uncertainty of which companies did or did not recover carbon, and if so, where it was subsequently sent. Additionally, if manganese was recovered, this was not included in the calculation due to the assumption of cobalt-containing cathodes. The results are shown in Table 6, along with efficiencies directly provided by recyclers.

Table 6. Recycling efficiencies

Company	Process	Location	Max calculated efficiency	Provided efficiency
P1	Pyrometallurgical	Europe	55.6%	64.9%
P2	Pyrometallurgical	Europe	31.1%	>65%
M1	Mechanical	Europe	69.6%	-
H1	Hydrometallurgical	Asia	65.3%	-
H2	Hydrometallurgical	North America	57.5%	-
H3	Hydrometallurgical	Asia	55.6%	-
C1	Combination	Europe	50.1%	-
C2	Combination	Europe	69.6%	52.2%

The difference between the calculated efficiency and provided efficiency is significant for all three companies that responded. The survey results indicated that Company P1 recovers energy from plastic incineration and uses recovered carbon as a reduction agent. It is likely that these were taken into account in the company's calculation, approximately accounting for the difference.

For Company P2, the maximum calculated efficiency is actually below the requirement set by the EU Battery Directive (50% recovery [17]). The difference can likely be attributed to the fact that Company P2 did not directly confirm their recovered materials. The survey results indicated that they do not recover steel and nickel. However, it is likely these metals are actually recovered to comply with the EU directive.

Unlike the other two recyclers, Company C2 provided an efficiency lower than the maximum calculated value. This company also provided their methods of calculation and it is clear that the difference in efficiency is primarily due to their calculations assuming 30% of plastic is recovered (as opposed to 100%, used in these calculations). Company C2 also used a different battery composition for their calculations.

Overall, there is a large range of estimated recycling efficiencies. The different between the companies can be primarily attributed to whether plastic was recovered, since it was assumed in these calculations that 100% of all plastic was recovered. If more accurate recovery efficiencies are taken into account, it is likely the difference would be smaller. Using the results shown in Table 6, on average, purely mechanical processes have the highest efficiency (~70%), followed by hydrometallurgical and combination processes (60%), with pyrometallurgical processes having the lowest efficiency (43%). The average efficiency for pyrometallurgical processes is greatly affected by the calculated efficiency for Company P2. Removing Company P2 from the analysis gives an average efficiency of 56% for pyrometallurgical processes.

3.4. Environmental impacts

The environmental effects of recycling lithium-ion batteries were evaluated in respect to the specific processes and the transport required between collection and recycling. A comparison was also made between the recycling of batteries and landfill.

In terms of the recycling processes, the survey results did not provide enough detailed information to calculate the environmental impacts directly. Therefore, a LCA was performed using secondary inventory data from 2004 [7] for both a hydrometallurgical and pyrometallurgical process. The inventory data was entered in GaBi LCA software and the effects on the three chosen impact categories were calculated. The results are shown in Table 7 and Table 8.

Table 7. Life cycle impact assessment, pyrometallurgical process

Process	GWP 100 (kg CO_2-eq)	HTP (kg DCB-eq)	TETP (kg DCB-eq)
Electricity generation	36.4	3.07	0.0891
Processing	0	0.0558	0
Plastics incineration	645	0.402	0.00499
Total	681	3.53	0.0941
Total (PE)	1.63e-11	1.37e-12	8.61e-14

Table 8. Life cycle impact assessment, hydrometallurgical process

Process	GWP 100 (kg CO_2-eq)	HTP (kg DCB-eq)	TETP (kg DCB-eq)
Electricity generation	16	1.36	0.0169
Processing	0	0.000783	9.87e-6
Landfill gypsum	817	0.754	0.493
Landfill residue	487	0.449	0.294
Total	1320	2.57	0.803
Total (PE)	3.16e-11	9.95e-13	7.35e-13

The results for the pyrometallurgical process indicate that the incineration of plastics has the largest impact on GWP 100 and electricity generation has the largest impact to HTP and TETP. For the analysis, a European distribution of energy sources was assumed in GaBi. However, the effects of electricity generation vary country to country and these effects could be reduced by implementing a larger proportion of energy generation from renewable sources. As for the incineration of plastics, the survey results have shown that it is not necessary to consume plastics in the heat treatment stage. Company P1 separated plastics mechanically before the heat treatment stage is performed.

The results for the hydrometallurgical process indicate that the landfill of gypsum and residue has the largest impact on GWP 100 and TETP, while electricity generation has the largest impact on HTP. The composition of the waste produced was not specified in the inventory data, so the impacts were modelled using a general landfill process. It may be possible to further treat the residue, resulting in less materials ending up in landfill.

Waste lithium-ion batteries are not currently processed in Australia. Therefore, there are environmental effects associated with their export. In order to make a general comparison of the transport to different continents, an analysis of the environmental effects was performed using LCA principles. Four general locations were chosen. It was assumed that the distance travelled by road was the same for each location, and was hence not included in the calculations. It was also assumed that batteries collected in Australia were shipped from Sydney. For the analysis, the transport option 'EU-27 – Container ship including fuel' was chosen in GaBi LCA software. The results are shown in Table 5.

The results show that the environmental effects of recycling batteries can be reduced by choosing recycling locations closer to Australia. For example, if batteries are recycled within Australia instead of being shipped to Europe, approximately 300kg CO_2-eq can be saved for each tonne of batteries transported. Furthermore, transporting batteries to Europe causes a 45% increase in GWP 100 impacts for pyrometallurgical processes, and a 550% increase in impacts to HTP for hydrometallurgical processes.

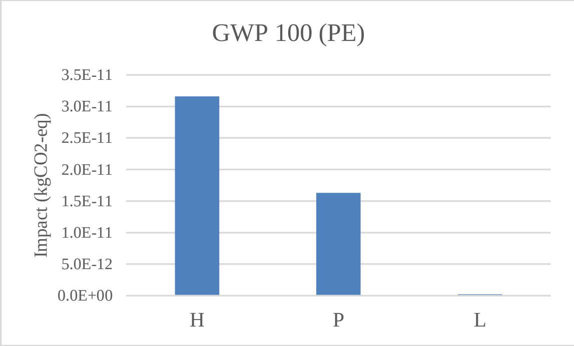

Figure 2. Comparison of processes in terms of GWP 100 impact

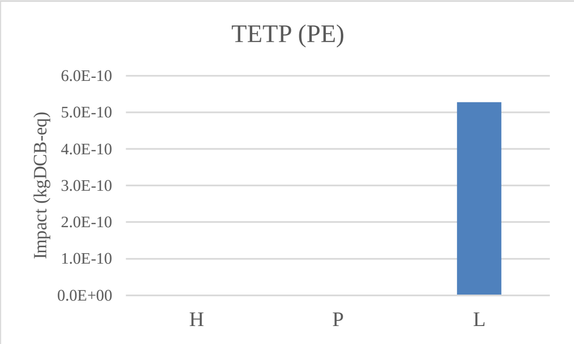

Figure 3. Comparison of processes in terms of TETP impact

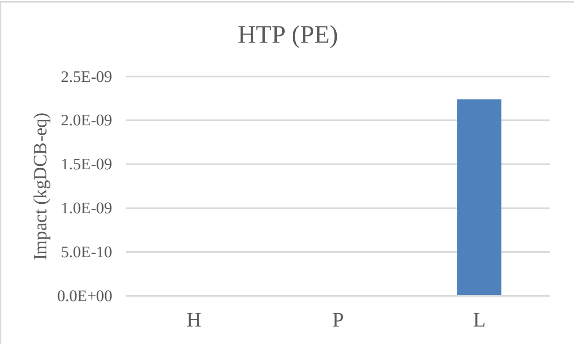

Figure 4. Comparison of processes in terms of HTP impact

To estimate the environmental effect of landfilling batteries, GaBi LCA software was used. Due to limitations in the software, only the impacts due to the nickel, copper and aluminium content of the batteries were assessed. The results were calculated assuming 5% of heavy metals were leached to soil [7]. It should be noted that the values for GWP 100 were not available through GaBi. Consequently, these values were calculated using emissions associated with landfill of mixed plastics and characterisation factors from the

Intergovernmental Panel on Climate Change [19]. The calculations for GWP considered only carbon dioxide, methane and nitrous oxide emissions from the plastic component of the battery. A comparison of all the LCA results is shown in Figure 2 to Figure 4.

For GWP 100, landfill showed a lower impact than the other processes. This result can be explained by the number of processes required for recycling, many of which involve carbon dioxide emissions. For both HTP and TETP, landfill showed a significantly higher impact when compared to recycling. Here, the effect on the environment is between three and four orders of magnitude higher when batteries are landfilled. It should be noted that these results take a conservative approach. The landfill estimations do not include several components of the batteries. Additionally, the recycling results do not take into account the negative impacts due to recycling, such as prevention of raw materials extraction.

4. Conclusions

The aim of this project was to investigate the different processes that are currently used for recycling lithium-ion batteries, and to compare these processes focusing on the associated environmental impacts. This information can be used by organisations such as the ABRI and MobileMuster to ensure the most appropriate disposal options are selected.

The results showed that the most commonly recovered materials are copper, nickel and cobalt, which correspond to the most valuable materials. It was found that hydrometallurgical processes recovered more materials than pyrometallurgical processes on average, with insufficient data to determine the number of materials recovered in purely mechanical processes. Of the eight companies surveyed, six claimed that plastic was recovered. However, of these six, only two companies showed plastic was further recycled, with the remaining companies either sending recovered plastic to landfill, consuming plastic in incineration processes with energy recovery, or not specifying the end process.

The life cycle assessment component of the study compared the environmental impacts between a hydrometallurgical and pyrometallurgical process, based on secondary life cycle inventory data. The results showed that for pyrometallurgical processes, the largest impacts are caused by plastics incineration for global warming potential, and electricity generation for human toxicity potential and terrestrial ecotoxicity potential. For hydrometallurgical processes, the largest impacts are caused by landfill for global warming potential and terrestrial ecotoxicity potential, and electricity generation for human toxicity potential.

The hydrometallurgical process showed a greater impact than both pyrometallurgy and landfill within the global warming potential impact category, while landfill showed the greatest impact for toxicity.

Transport of waste batteries for processing was also found to have a significant effect on the overall impact. For example, transporting batteries from Australia to Europe was found to increase the global warming potential by 45% for pyrometallurgical processes, and the human toxicity potential by 550% for hydrometallurgical processes.

The results overall show that to decrease the environmental impacts of recycling portable lithium-ion batteries, processes that utilize low temperatures and are capable of recovering plastic should be used. Furthermore, the impacts can be decreased by reducing the distance travelled between collection and recycling.

Acknowledgements

The authors would like to express thanks to the Australian Battery Recycling Initiative and MobileMuster Australia for providing guidance in this study.

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