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PRELIMINARY INVENTORY OF ALKALINE BATTERIES LANDFILLING

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ABSTRACT

Life cycle assessment (LCA) is a technique for assessing the environmental impact of a system or product from cradle to grave, i. e. from the extraction of raw materials to the final disposal. The need of including waste treatment step in a product life cycle analysis and the wide use of this tool to evaluate different options on solid waste management practices had increased the attention of LCA practitioners for this final step of the products life.

In the case of alkaline batteries - a complex product that contains metal, plastic and mineral fractions, including heavy metals, this analysis is impeded by the lack of scientific data concerning its behavior when incinerated or disposed of in a landfill.

Under this subject, a project to compare the environmental impact of incineration, landfilling and recycling of alkaline batteries using LCA is undergoing at the Engineering Faculty of Porto University.

The objective of this paper is to present and discuss the preliminary inventory of batteries landfilling in terms of consumption of materials and energy, emissions to air, water and solid waste. This inventory was performed in two parts. On the first one a theoretical model was used to estimate resources and emissions based on batteries characterization. On the second one, results from leaching laboratorial experiments made with the target batteries were compared with the theoretical model approach.

The result of this preliminary inventory shows the potential contribution of landfilled batteries to the environmental burdens of a landfill and also allows an evaluation of the relative contribution of the different batteries components to these burdens.

KEYWORDS

Batteries; inventory; leaching; landfilling; life cycle assessment.

INTRODUCTION

The degradation process in a landfill where degradable organic materials are present, e.g. in a landfill for municipal solid waste (MSW), can be described as a sequence of different phases. The initial phase concerns the time period from the opening of the landfill until the biological degradation accelerates. In the second still aerobic phase, the degradation of easily degradable organic material takes place, leading to carbon dioxide generation. This phase is followed by the acid anaerobic phase resulting in the generation of fatty acids, carbon dioxide and some hydrogen. The solubility of many metals increases during this phase. The fourth phase, or the methanogenic phase, is an anaerobic process characterized by methane production. In the fifth phase, called the maturation phase, only the more stable components remain and the methane production slows down. The sequence described here applies to a homogeneous waste volume. A landfill will be composed of cells of highly varying age and composition, and different parts of the landfill can therefore be at different phases of degradation (Christensen, 1989).

Emissions from landfills may prevail for a very long time, often thousand years or longer (Finnveden et al., 1995). When dealing landfilling in connection with LCA, two difficult questions arise: the time period in which emissions should be integrated and the allocation of inputs and outputs to the product under study. In a product LCA we usually want some kind of causal relation between the studied product or material and each emissions actually caused by it. However, presently available data only shows the total emissions. Furthermore, it is not possible to measure emissions from certain products. The causal relations must be calculated by some kind of model, to partition in some proportionate shares the responsibility for environmental impacts caused by different inputs (Sundqvist, J.- O., 1999). This has at least two implications: (i) the emissions from a landfill must be predicted and, therefore, it is the potential emissions rather than actual emissions that can be included in the product LCA for the landfill process; (ii) a choice concerning which time is of interest must be taken in order to make the potential emissions from landfilling comparable to other emissions (Finnveden et al., 1995).

The theoretical model used on the first part of this inventory is the one proposed in Sundqvist, J.-O., 1999 and presented in several reports (Sundqvist et al., 1994, Finnveden et al., 1995, Finnveden and Huppes, 1995, Finnveden, 1996 and Sundqvist et al, 1997). On this model, the potential emissions are estimated from the elemental and chemical composition of the material. The time influence is based on two time horizons, respectively a shorter and a longer time period. The shorter time period is based on the degradation processes in the landfill and is called surveyable time period. It is defined as the time period until the landfill reaches some kind of pseudo-steady-state. It should be of the magnitude of one century. For MSW landfills, the surveyable time period was identified as the period until the later part of the methanogenic phase. The longer time period is called the hypothetical, infinite time, when all landfilled material has been released to the environment. It is also a worst-case scenario (Sundqvist, J.-O., 1999). For both cases, the surveyable time period and the hypothetical infinite time period, the emissions are assumed to occur at the current moment.

METHOD OF APPROACH

The methodology followed on the development of this work has the following steps:

- 1. batteries characterization
- 2. preliminary inventory of batteries landfilling using the theoretical model
- 3. laboratory leaching experiments with batteries

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Batteries characterization

The first step on this work was characterizing the target batteries. Batteries are complex products containing metals, plastics and mineral fractions, including heavy metals.

Studied batteries are alkaline zinc-manganese dioxide batteries, commonly referred to as alkaline batteries (Hurd et al., 1993). These batteries account for the major market share of household batteries in Portugal and even in the entire world. The most popular format is the AA standard, a cylinder with 50mm length, 14mm diameter and approximately 23.5g of weight. Alkaline batteries use powdered zinc as the negative electrode (anode), manganese dioxide (MnO₂) with graphite as the positive electrode (the cathode), and highly conductive potassium hydroxide (KOH) as the electrolyte (Hurd. et al., 1993). A cylindrical steel can is the container for the cell that also serves as the cathode current collector. The cathode is positioned inside the can in the form of a hollow cylinder in close contact with the can inner surface. Inside the hollow center of the cathode are placed layers of paper and cellophane as separator. Inside of that is the anode, made from powdered zinc, with a collector of brass contacting it, and making connection through a plastic seal to the negative terminal of the cell. The cell has a top cover and a plastic sleeve applied (modified from Linden, D., 1995).

Data concerning the batteries characterization and composition used on the present study are based on the results of an exhaustive laboratory work (Xará et al., 2002a) and are succinctly presented on Table 1.

Table 1. Characterization of spent alkaline batteries analyzed on the present study

Material/Element	Average dry weight (g/battery)	Trace elements	Average amount (mg/battery)
Ctool	4.7		
Steel	4.7	As	0.021
Tin-plated brass	0.44	Cd	0.060
PVC	0.23	Со	0.84
PA	0.22	Cr	9.5
Cardboard	0.060	Cu	281
Paper	0.11	Hg	0.0038
Cellophane	0.045	Mn (except cathode)	7.0
Mn (on cathode)	5.4	Ni	65
C (on cathode)	0.71	Pb	1.2
Zn (on anode)	3.4	Sb	0.44
КОН	0.92	Si	0.84
Others (including oxygen and water)	7.3	TI	1.4
		V	0
		Zn (except anode)	218

Preliminary inventory of batteries landfilling using the theoretical model

In the theoretical model a conventional MSW landfill where mixed waste materials are deposited was considered. Wastes enter the landfill site in the collection or transfer vehicles, are spread out and then compacted. The waste deposited in each operating period, usually one day, forms an individual cell (Tchobanouglous et al, 1993) that is considered daily covered with soil to protect the waste from birds and rats and to avoid odors. When the landfill is finished a final cover is applied.

Besides waste, the inputs in the process consist of soil for covering the waste and diesel oil for the compactor. The energy consumption of the compactor is assumed as 1 liter of diesel oil per ton, equivalent to 40 kJ/ton. On Sundqvist, J.-O., 1999, it is referred that the allocation of diesel consumption can be discussed, having at least two possibilities in accordance with the causality principle, respectively allocation by volume or by weight. On this work the allocation by weight was considered. Soil consumption was assumed as $0.08 \, \mathrm{m}^3$ /ton of waste for the daily cover and $0.125 \, \mathrm{m}^3$ /ton of waste for the final cover.

The landfilling process itself involves the decomposition of part of the landfilled waste. The outputs from the process are the final stabilized solid waste plus the gaseous and aqueous products of the decomposition, which emerge as landfill gas and leachate (White et al, 1995). Different substances degrade with different rates. The degradation yield during the surveyable time period considered for the organic materials of batteries was 3% for PVC and PA and 70% for paper, cardboard and cellophane components. From the chemical formulae of these components it is possible to estimate emissions of CH₄, CO₂, and NH₃ by the following general mechanism of organic material degradation:

$$C_{a}H_{b}O_{c}N_{d}S_{e} + (a - \frac{b}{4} - \frac{c}{2} + \frac{3d}{4} - \frac{e}{2})H_{2}O \longrightarrow (\frac{a}{2} + \frac{b}{8} - \frac{c}{4} - \frac{3d}{8} - \frac{e}{4})CH_{4} + (\frac{a}{2} - \frac{b}{8} + \frac{c}{4} + \frac{3d}{8} + \frac{e}{4})CO_{2} + dNH_{3} + eH_{2}S$$
(1)

In the case of PVC, it was also considered that all the Cl on the degraded portion of the material produces HCl.

The model considers that 1% weight of the degraded carbon will outflow via leachate mainly as fatty acids and 99% via the landfill gas as CH₄ and CO₂. As a thumb rule, 1g of total organic carbon (TOC) corresponds to 3g of chemical oxygen demand (COD). The average ratio BOD/COD is assumed to be 0.25 during the surveyable time period. On the infinite time period a complete degradation of the materials occurs and the major degradation product is assumed to be CO₂ (Sundqvist et al, 1997).

The landfill considered has gas extraction and leachate collection systems with gas collection efficiency assumed as 50% of the amount produced. The collected gas is flared on the site burning the combustible fractions and destroying most of the organic trace components. The methane emission is also reduced by methane oxidation on the soil cover and 15% of the amount not collected was assumed oxided in such circumstances. The leachate is collected and usually treated, however emissions from its treatment are not considered here due to lack of data.

Metals in MSW can be divided into metallic materials and metal compounds as for example additives in plastics and paper. Before the metals can be emitted via leachate they must be

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released from the solid waste matrix into the percolating water. The metals emitted from batteries from both metal components and trace elements on other components were based on the emissions factors presented on the theoretical model used.

The results obtained for this preliminary inventory of batteries expressed for one battery unit are presented on Table 2.

Laboratory leaching experiments with batteries

The leaching laboratory experiments were carried out in four test columns filled with batteries, either entire or cross-cut, that were continuously leached with two different solutions, either deionised water or nitric acid solution (pH4). Samples were periodically taken and heavy metals determined. The general methodology followed on these tests is based on the standard NEN 7343. The results of the total amount of each metal leached at the end of 25 days (columns 1 and 2) and 21 days (columns 3 and 4) are presented on Table 3. Detailed results from these experiments are available on Xará et al., 2002b.

Table 2. Preliminary inventory of batteries landfilling based on theoretical calculations

Inputs		
Energy (I of diesel/battery)	2.35E-05	
Soil cover (m³/battery)	4.82E-06	
Outputs	STP	ITP
Air emissions (mg/battery)		Ŷ.
CO ₂	195.7	1078
CH₄	19.9	19.9
HCI	, 3.9	3.9
NH ₃	0.96	0.96
Cd	3.00E-06	6.00E-03
Hg	1.90E-07	1.90E-03
Water emissions (mg/battery)		
TOC	0.69	0.69
COD	2.1	2.1
BOD	0.52	0.52
As	4.20E-05	2.10E-02
Cd	2.70E-05	5.40E-02
Cr	6.65E-03	9.50E+00
Cu	1.97E-02	2.81E+02
Fe	4.66E-01	4.66E+03
Hg	1.90E-07	1.90E-03
Ni	3.25E-01	6.50E÷01
РЬ	7.20E-05	1.20E+00
Zn	7.14E-01	3.57E+03

STP = surveyable time period; ITP = infinite time period

Table 3. Results of leaching tests performed with spent alkaline batteries

Metal	Entire batteries nitric acid solution 25 days	Entire batteries deionised water 25 days	Cross-cut batteries nitric acid solution 21 days	Cross-cut batteries deionised water 21 days
	Amount leached (mg	/battery, except As a	and Hg expressed as p	ıg/battery)
As	<ld< td=""><td><ld< td=""><td>0.0242</td><td>0.0217</td></ld<></td></ld<>	<ld< td=""><td>0.0242</td><td>0.0217</td></ld<>	0.0242	0.0217
Cd	<ld< td=""><td><ld< td=""><td>0.0001</td><td><ld< td=""></ld<></td></ld<></td></ld<>	<ld< td=""><td>0.0001</td><td><ld< td=""></ld<></td></ld<>	0.0001	<ld< td=""></ld<>
Со	<ld< td=""><td><ld< td=""><td><ld< td=""><td><ld< td=""></ld<></td></ld<></td></ld<></td></ld<>	<ld< td=""><td><ld< td=""><td><ld< td=""></ld<></td></ld<></td></ld<>	<ld< td=""><td><ld< td=""></ld<></td></ld<>	<ld< td=""></ld<>
Cr	0.0130	0.0083	0.0017	0.0013
Cu	<ld< td=""><td><ld< td=""><td>0.0058</td><td>0.0067</td></ld<></td></ld<>	<ld< td=""><td>0.0058</td><td>0.0067</td></ld<>	0.0058	0.0067
Fe	0.1897	0.0653	0.0493	0.0075
Hg	<ld)<="" td=""><td><ld< td=""><td>0.2156</td><td>0.1001</td></ld<></td></ld>	<ld< td=""><td>0.2156</td><td>0.1001</td></ld<>	0.2156	0.1001
Mn	0.0021	0.0008	0.0491	0.0235
Ni	0.0266	0.0001	0.0102	0.0153
Pb	<ld< td=""><td><ld< td=""><td>0.024</td><td>0.0072</td></ld<></td></ld<>	<ld< td=""><td>0.024</td><td>0.0072</td></ld<>	0.024	0.0072
Sb	<ld< td=""><td><ld< td=""><td><ld< td=""><td><ld< td=""></ld<></td></ld<></td></ld<></td></ld<>	<ld< td=""><td><ld< td=""><td><ld< td=""></ld<></td></ld<></td></ld<>	<ld< td=""><td><ld< td=""></ld<></td></ld<>	<ld< td=""></ld<>
Tl	<ld< td=""><td><ld< td=""><td><ld< td=""><td>0.0008</td></ld<></td></ld<></td></ld<>	<ld< td=""><td><ld< td=""><td>0.0008</td></ld<></td></ld<>	<ld< td=""><td>0.0008</td></ld<>	0.0008
Zn	0.0092	0.0350	8.9301	9.7397

<LD = below the detection limit

DISCUSSION AND CONCLUSIONS

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The theoretical model results for estimating emissions from alkaline batteries landfilling assuming the two different time periods, show meaningful lower emissions of CO₂ from the organic fractions as well as metals on the surveyable time period than on the infinite time period. Therefore, according to the model, most of the emissions will occur after the surveyable time period. This conclusion is expressive for all metals where Zn, Fe and Ni show the highest release on the surveyable time period.

Results from the leaching tests show that metals are more strongly leached from cross-cut batteries than the entire ones, except for chromium and iron, and nickel with nitric acid solution. The larger difference between metals released from entire and cross-cut batteries is for Zn that shows appreciable values only for cross-cut batteries where anode is exposed to the leaching solution.

On entire batteries the extraction with nitric acid solution was higher than with deionised water for Cr, Fe, Mn and Ni, but surprisingly lower for Zn. It seems that Zn acts as a cementation agent for the other metals most of which from the external steel can. With the cross-cut batteries this difference is sensible for Fe, Hg and Pb. Again for Zn, extraction with deionised water on cross-cut batteries was higher than with nitric acid solution.

The metal emissions estimated by the theoretical model on the surveyable time period are less than those in the leaching tests on the cross-cut batteries for Hg, As, Pb, Zn and Cd. On the contrary, Cu, Fc and Ni emissions are over-estimated by the theoretical model. The differences

are quite meaningful Pb and Zn 100 times

Such discrepancies conditionates its reludeionised water are e leached must not be not equal, and, clearly

Finally, whether or n entire batteries are amounts of metals.

ACKNOWLEDGEN

Authors acknowledge the research project P

Susana Xará acknow the program Praxis X

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od are less Cd. On the are quite meaningful for Hg and As, respectively in the range of 10⁶ and 10³ times lower, and for Pb and Zn 100 times less.

Such discrepancies allow us to conclude that the state of the metals on batteries strongly conditionates its release and general rules fail. Anyway, neither the nitric solution nor the deionised water are expected to be the leachant at the MSW landfill, thus the amounts of metals leached must not be the same. Moreover, the oxidation potential of both types of solutions are not equal, and, clearly, 21 days is not the same as 100 years.

Finally, whether or not the batteries are damaged strongly influences the metals released. When entire batteries are landfilled they must be seriously corroded before releasing significant amounts of metals.

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Xará, S., Silva, M., Almeida, M.F. and Costa, C., Laboratory study on the behavior of spent alkaline batteries in landfilling and incineration Part 2 – Batteries leaching, manuscript, 2002b.

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ABSTRA(

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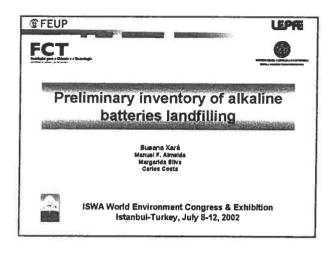
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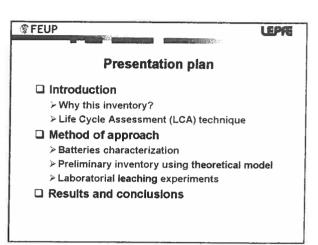
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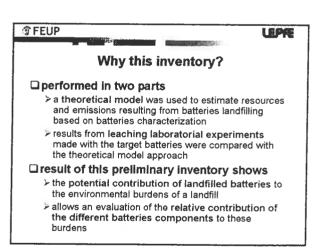




Why this inventory? □ part of a project ➤ to compare the environmental impact of incineration, landfilling and recycling alkaline batteries using Life Cycle Assessment (LCA) technique □ present and discuss the preliminary inventory of batteries landfilling ➤ in terms of consumption of materials and energy, emissions to air and water and, solid waste

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Life Cycle Assessment (LCA)

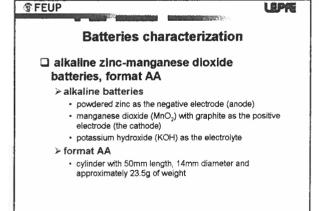
- ☐ Technique for assessing the environmental aspects and potential impacts associated with a product by:
 - Compiling an inventory of relevant inputs and outputs of a product system
 - > Evaluating the potential environmental impacts associated with those inputs and outputs
 - Interpreting the results of the inventory analysis and impact assessment phases in relation to the objectives of the study

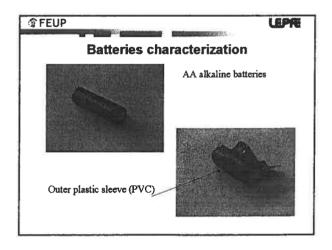
Product system

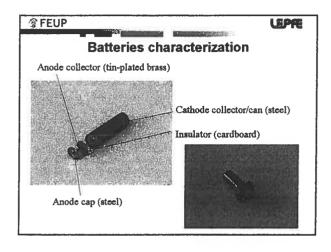
Collection of materially and energetically connected unit processes which performes one or more defined functions

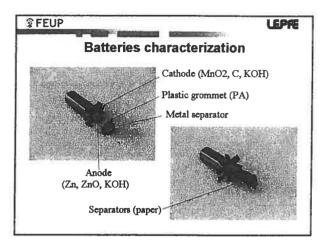
Method of approach

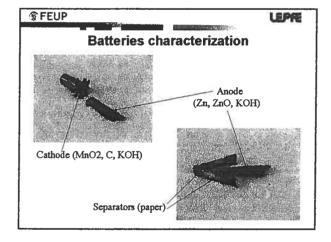
- 1. Batteries characterization
 - Strutural components
 - · Chemical composition
- 2. Preliminary inventory of batteries landfilling using a theoretical model
 - Landfill characterization
 - · Landfilling process modelling
- 3. Laboratorial leaching experiments
 - Tests description

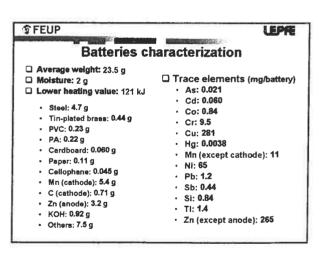


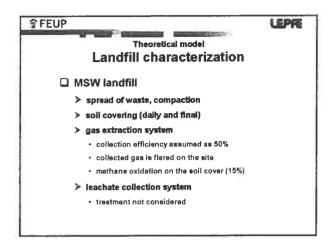


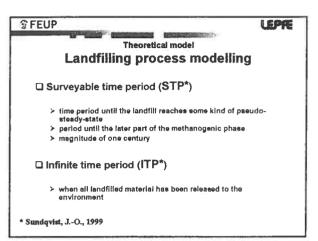


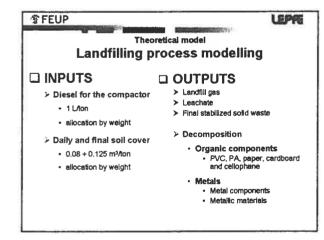






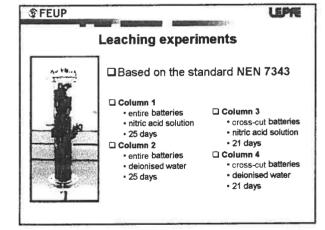






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Emiss	ion factor fo	r metals	kg emitted/kg landfilled)
	STP	ITP	
As	2 x 10-3	1	leachate
Cd	5 x 10-4	1	10% gas, 90% leachate
Cr	7 x 10-4	1	leachate
Cu	7 x 10-5	1	leachate
Fe	1 x 10-4	1	leachate
Hg	1 x 10-4	1	50% gas, 50% leachate
Ni	5 x 10 ⁻³	1	leachate
Pb	6 x 10-4	1	leachate
Zn	2 x 10-4	1	leachate

♀FEUP			LEPPE
		1000	
Theoretical model	Outputs Air emissions (mg)	STP	ITP
Results (1 battery)	co.	195.7	1078
	CH.	19.9	19.9
Inputs	HCI	3.9	3.9
•	NH ₂	0.96	0.96
Diesel (L): 2.35 x 10*	Cd	3.005-04	6.60H-03
Soil cover (m³): 4.82 x 104	Hg Water emissions (mg)	1.90至-07	1.86E-03
	TOC	0.69	0.69
	COD	2.1	2.1
	BOD	0.52	0.52
	As	4.20E-05	2.10H-02
	TOC 0.1 COD 2. BOD 0.1 As 4.28 Cd 2.76	2.70⊞-05	5.40E-02
	Cr	6.65E-03	9.56E+00
	Cu	1.87E-02	2.815+02
	Fe	4.66E-01	4,66E+03
	Hg	1.905-67	1.906-03
	Ni	3.25E-01	6.5026+01
	Pb	7.20E-05	1,202+00
	Z n	7.148-01	3.57E+03



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Le	aching exp	eriments	- Results (battery)
Metal	Entire batteries nitric scid solution 25 days	Entire batteries delonised water 26 days	Cross-cut batteries nitric sold solution 21 days	Cross-cut batteries delonised water 21 days
A	mount leached (mg/t	attery, except As	and Hg expressed a	: Tg/bzttery)
As	< L,D	< LD	0.0242	0.0217
Cd	< LD	< LD	8.0001	< LD
Co	< LD	< LD	< LD	< LD
Cr	0.0130	0.0003	0.0017	0.0013
Çu	∢LD	< LD	0.0058	0.0067
Fe	0.1997	0.0683	0.0493	0.0076
Hg	< LD	< LD	0.2166	0.1001
Mn	0.0021	0.0008	0.0491	0.0236
NI	0.0286	0.0001	0.0102	0.0163
Pb	< LD	< LD	0.024	0.0072
Sb	< LD	< LD	< LD	< LD
П	< LD	< LD	< LD	0.0008
Zn	0.0082	0.0350	8,9301	9.7307

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Results and conclusions

☐ Theoretical model

- > most of the emissions (CO2 from the organic fraction and metale) will occur after the surveyable time period
 this conclusion is expressive for all metals where Zn, Fe and Ni show the highest raisease on the surveyable time period

☐ Leaching tests

- cross-cut vs. entire

 - cross-cut vs. entire
 metals are more strongly leached from cross-cut batteries than the entire ones,
 except for Cr and Fe, and Ni with nitric acid solution
 the larger difference between metals released from entire and cross-cut batteries
 is for Zn that shows appreciable values only for cross-cut batteries where anode
 is exposed to the leaching solution
- intric acid solution vs. delonised water
 on entire batteries the extraction with ritio acid solution was higher than with delonised water for Cr, Fe, Mn and NI, but lower for Zn
 with the cross-cut batteries this difference is sensible for Fe, Hg and Pb. Again for Zn, extraction with delonised water on cross-cut batteries was higher than with hitric acid solution.

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Results and conclusions

- ☐ Theoretical model vs. Leaching tests
 - Lower emissions estimated by theoretical model
 Crose-cut batteries: Hg, Ae, Pb, Zh and Cd
 Higher emissions estimated by theoretical model
 Cu, Fe, Ni

 - > the state of the metals on batteries strongly conditionates its release
 - neither the nitric solution nor the delonised water are expected to be the leachant at the MSW landfill, thus the amounts of metals leached could not be the same
 - the oxidation potential of both types of solutions are not equal, and, clearly, 21 days is not the same as 100 years
 - whether or not the batteries are damaged strongly influences the metals released
 - when entire batteries are landfilled they must be seriously corroded before releasing significant amounts of metals.

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