Development of a Calculation Method for Recycling Efficiencies

of Battery Recycling Processes

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1 Introduction

In July 2006, the European Parliament and the EU Council of Ministers agreed on a compromise to revise the 1991 Directive covering batteries and accumulators. The new Directive provides (inter alia) for recycling and collecting targets to be reached by 2011 and 2012 respectively at the latest. It has been set that any process for the recycling of batteries will be obliged to reach a recycling efficiency of 65 % by average weight for lead-acid batteries, 75 % for nickel-cadmium and 50 % for other battery types. Due to various reasons the recycling efficiency of a process cannot be measured reliably, so that it has to be calculated. But while the recycling targets have been fixed, it has not been defined how the recycling efficiency is calculated. Up to now no agreement on a calculation method for recycling efficiencies regarding battery recycling processes have been finalized. However the calculation method will have a major impact on the battery recycling industry in Europe. Critical points that have to be reflected by an equation are for example the definition of system boundaries, of battery elements and components to be considered and those to be neglected, of assigning material values to the input and output materials and of ensuring a transferability of the equation to all related battery systems.

This paper is based on a study at IME Aachen with the objective to develop a simple calculation method which can be applied to the recycling processes applied nowadays in industry and which does not favour any processing route in particular. But the more simplified the method is the more compromises are to be accepted as it will be shown. The method presented here is offered as a discussion basis for the European expert group of all stakeholders, which have to come to a mutual agreement within the near future.

2. Calculation Basis for Recycling Efficiencies

2.1 Definition of Recycling

Before going into details on recycling efficiency the definition of recycling in the case of batteries has to be discussed. According to the EU Battery Directive the recycling of batteries means the "processing of waste batteries and accumulators for generation of products that can be directly reused in battery production or in other applications or processes". The definition excludes the possibilities of disposal or energy recovery. Unfortunately a repair and reuse of waste batteries is impossible. Thus the only way of recycling is the recovery of value materials (not only metals). That leads to the question which kind of materials can be considered for the calculation. Waste batteries may contain a lot of materials which were generated by different chemical reactions during the use of the batteries. Those reaction products were not present in new batteries and it is not possible to analyse all of them in the battery scrap. Hence we propose to take only materials used for manufacture of new batteries into account for the calculation. Such materials are metals, metal salts and oxides, carbon, plastics, electrolyte or papers. An advantage of this method is that these materials are well-known in composition and weight fractions because of the material data sheets that exist for all types of batteries.

2.2 Key Elements for the Calculation

For the proposed simple calculation method the following key elements are set as the calculation basis for recycling efficiencies:

1. prescribed recycling efficiencies apply for the average weight of the battery scrap
2. all elements or compounds being present in new batteries that can be captured and clearly identified in a product can be taken into account
3. elements or compounds originating from new batteries that are chemically or energetically used during the recycling process do not count according to the directive
4. as reference for calculating the recycling efficiencies the mass of the waste batteries without humidity is used (according to the EU Battery Directive), not considering any plastic shells or electronics from battery packs
5. recycling efficiency is defined as the weight ratio of the acceptable “product fraction(s)” and the considered battery scrap mass
6. battery recycling processes may consist of multiple (sub)steps; each step is assessed individually and recovery values have to be added
7. any material produced during the entire recycling process that cannot be considered as a product with regard to the EU Waste Directive is not accepted
8. all produced elements and downstream compounds are to be accepted by their full weight, if this element or compound was component of the new battery

2.3 Average Weight

Figure 1 shows a process example for explanation of the “average weight”. Starting from a collected mix of waste batteries containing all types (100 % input) you may get different battery fractions after sorting. In this exemplary 30 % of fraction A, 60 % of fraction B and 10 % of fraction C is assumed.

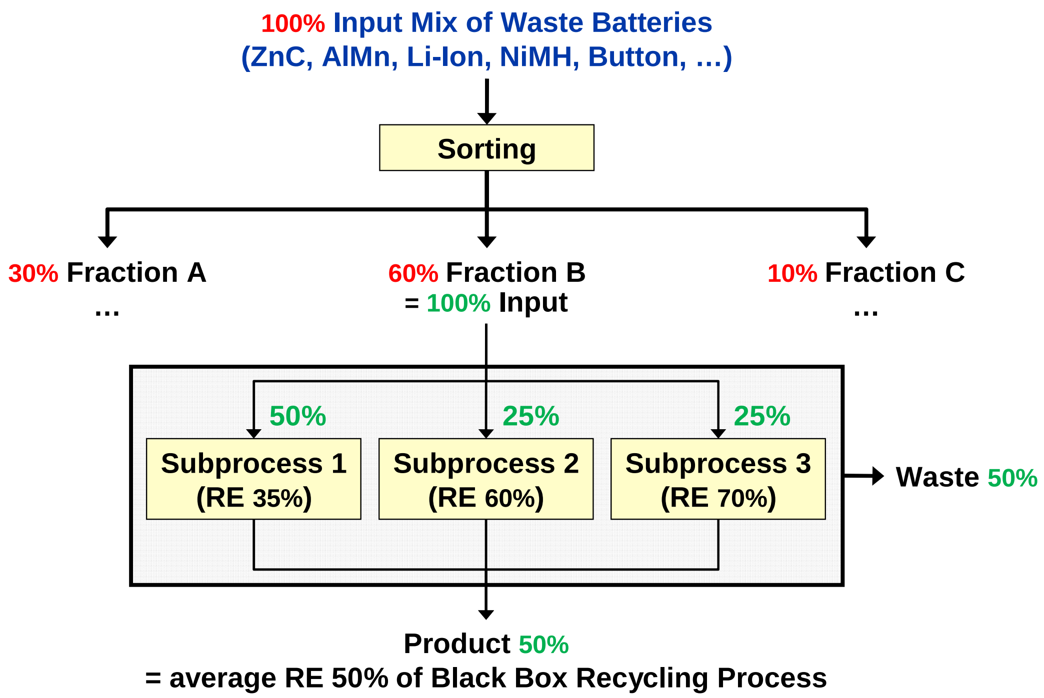


Figure 1: Process example for explanation of “average weight”

Fraction B may contain no lead-acid or nickel-cadmium batteries from the input mix, and should represent 100 % input of a recycling process. This might be considered as a black box with an overall recycling efficiency of 50 %, but also might be divided into three different sub processes with differing recycling efficiencies of 35 % for sub process 1, 60 % for sub process 2 and 70 % for sub process 3. Even though sub process 1 does not fulfil the prescribed recycling efficiency of 50 %, it is to be accepted. Not the single sub process is to be considered but the average recycling efficiency of the total (“black box”) recycling process has to fulfil the prescribed recycling efficiency. This procedure may allows for economical optimisation, e.g. for the case that sub processes 2 and 3 are more expensive than sub process 1. The case will be become more complex to control, if sub processes are run in different countries. In this example the black box recycling process fulfils the EU guidelines for fraction B, but the total input mix of waste batteries have to be investigated accordingly.

2.4 Definition of Product

Following chapter 2.2 the recycling efficiency is defined as the weight ratio of acceptable "product fraction(s)" and the considered battery scrap mass. Thereby acceptable products should be defined as follows:

- end-products or feedstock in other processes
- materials not classified as waste with regard to the EU Waste Directive
- materials independent of market value or final use

2.5 The Specific Case of Water, Oxygen, Carbon and Slag

Water

For calculation of the recycling efficiency only the mass of the battery cell without humidity counts (s. EU Battery Directive, Annex III, Detailed Treatment and Recycling Requirements, Part A: Treatment), so water is not considered at all.

Oxygen

Oxygen can only be taken into account for the calculation if it is part of a compound in an approved recycling product and this compound (formula) was already present as component in the virgin battery (e.g. MnO_2, NiOOH, $LiCoO_2$...).

Carbon

Only if carbon is part of the new battery and is unchanged present in an approved recycling product, it can be taken into account (e.g. graphite powder, FeMnC...). So if a recycler extracts the carbon from the battery scrap, it counts as product, but if the carbon is used in the process as a reducing agent, it is not considered.

Slag

If the slag is an approved product according to the EU Waste Directive (e.g. used for road or dump construction), the content of battery relevant metal or metal oxides can be used for the recycling efficiency calculation. But if the slag is declared as waste and described to landfill, it cannot be taken into account for the calculation of the recycling efficiency.

3. Battery Recycling Processes

3.1 Fundamental Differentiations

A variety of battery recycling processes exist which can be divided into pyrometallurgical and hydrometallurgical processes (s. Figure 2 and 3). Pyrometallurgical processes primarily aim on the high rate recovery of metals at high temperatures, but often with limited selectivity using smelting furnaces whereas hydrometallurgical processes aim on the selective recovery of elements or compounds at low temperatures by dissolving and precipitation, but typically with lower $t/(h{\cdot}m^3)$ rates). Some battery recycling processes combine pyro- and hydrometallurgical steps and often have pre-treatment steps like pyrolysis or mechano-physical cell processing, i.e. crushing and material separation as shown in Figure 4.

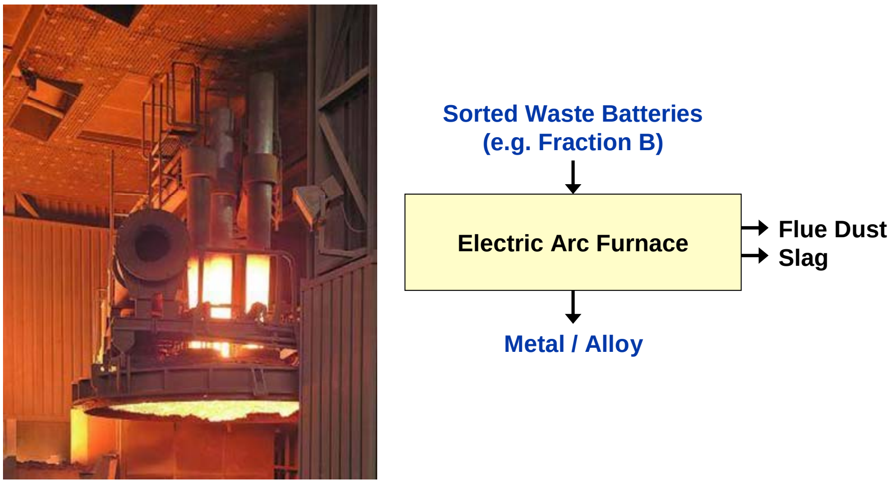


Figure 2: Example for a pyrometallurgical battery recycling process

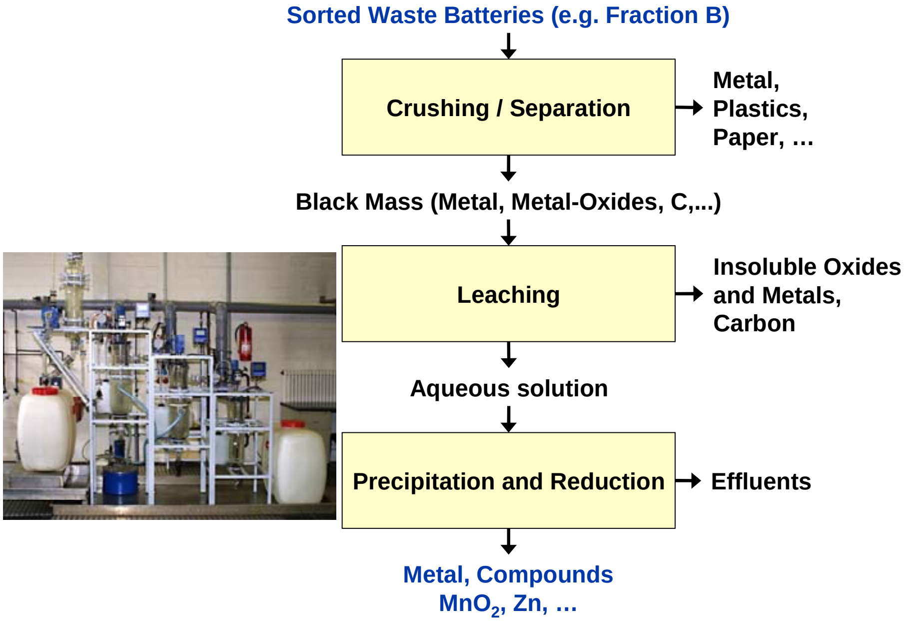


Figure 3: Example for a hydrometallurgical battery recycling process

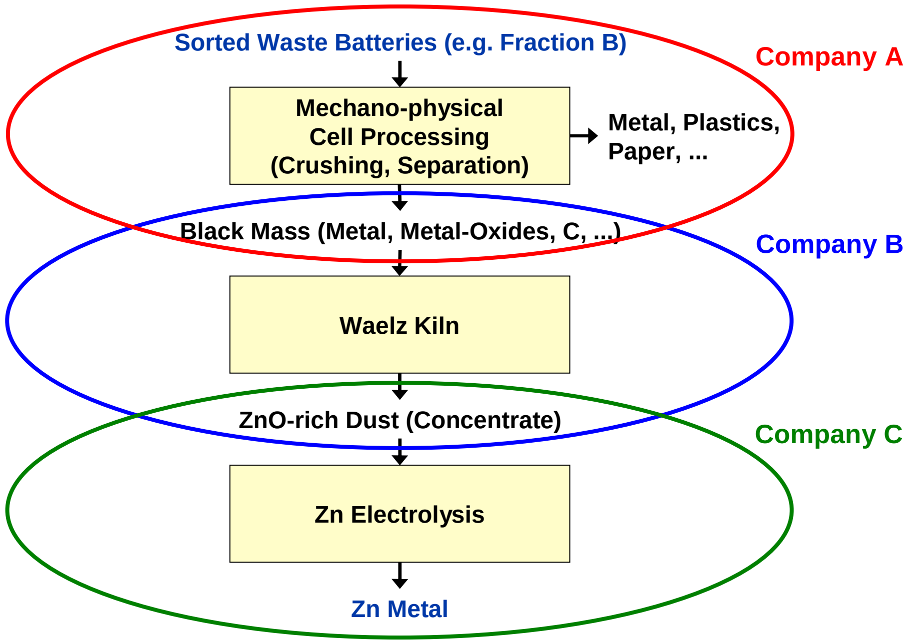


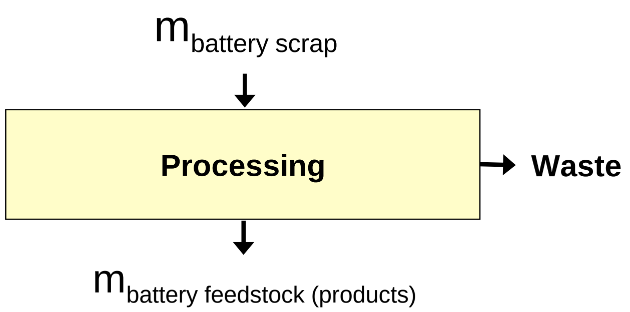
Figure 4: Example for a battery recycling process that combines pyrometallurgical and hydrometallurgical process steps at different sites (incl. pre-treatment)

Besides utilisation of specialised battery recycling processes the addition of waste batteries to non battery dedicated large-scale processes (e.g. extractive zinc, cobalt or nickel metallurgy) is common practise and very often an economical advantage. This case has also to be considered by the efficiency calculation methodology without making it too complex as economy besides ecology is a strong aspect of sustainability.

4. Calculation Method for Recycling Efficiencies

4.1 Closed Loop Technology

Closed loop technology means that the products generated within the battery recycling process can be directly used for the production of new batteries again. For closed loop recycling processes the calculation of the recycling efficiency (RE) is the simplest case, as RE is equal to the weight ratio of all approved products and the input battery scrap mass (s. Figure 5).



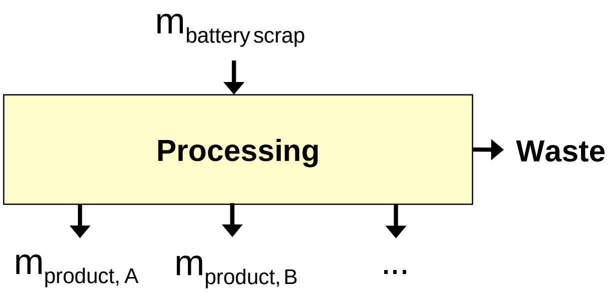
$$RE^{total}: \quad RE_{battery} = \frac{m_{products}}{m_{battery\ scrap}}$$

Figure 5: Calculation of recycling efficiency for closed loop technology

4.2 Open Loop Technology (Dedicated Process)

Open loop technology means that the products generated within the battery recycling process can be used for all kind of production or product manufacturing as feedstock materials. In the case of open loop technology processes dedicated to battery recycling the recycling efficiencies of each battery element (e.g. metal) and each compound (e.g. metal oxide) have to be considered. When an element or compound X is contained in more than one product (s. Figure 6) the mass fractions of X in every product have to be added and the sum is divided by the input battery scrap mass. This leads to the recycling efficiency of X.

To calculate the total recycling efficiency all individual recycling efficiencies of X, Y… have to be summed up.



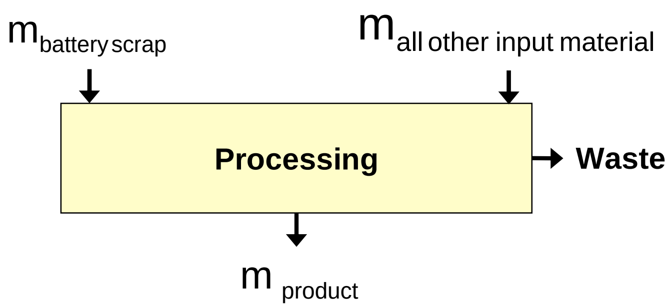
$$RE^X: \quad RE^X_{battery} = \frac{m^X_{product,A} + m^X_{product,B} + \dots}{m_{battery\,scrap}} \quad \text{mit} \quad m^X_{product,i} = m_{product,i} \cdot c^X_{product,i}$$

$$RE^{total}: \quad RE_{battery} = RE^X_{battery} + RE^Y_{battery} + \dots$$

Figure 6: Calculation of recycling efficiency for open loop technology (dedicated processes)

4.3 Open Loop Technology (Add to Process)

If waste batteries are added to non battery dedicated large-scale processes, a factor has to be introduced, considering that the battery scrap mass is not the only input low for battery relevant element or compound X. In this case simply the total input mass of X has to be adjusted by factor K whereas K is the weight ratio of X in the battery scrap and the total input mass of X. Then the individual battery recycling efficiency of X is equal to the ratio of mass fraction of X in the product and the input battery scrap mass multiplied by K. If more than one product can be considered, the formulas have to be extended accordingly as discussed in the previous chapter 4.2.



$$RE^X: \quad RE^X_{battery} = K^X \cdot \frac{m^X_{product}}{m_{battery\ scrap}} \quad \text{mit} \quad K^X = \frac{m^X_{battery}}{m^X_{battery} + m^X_{others}}$$

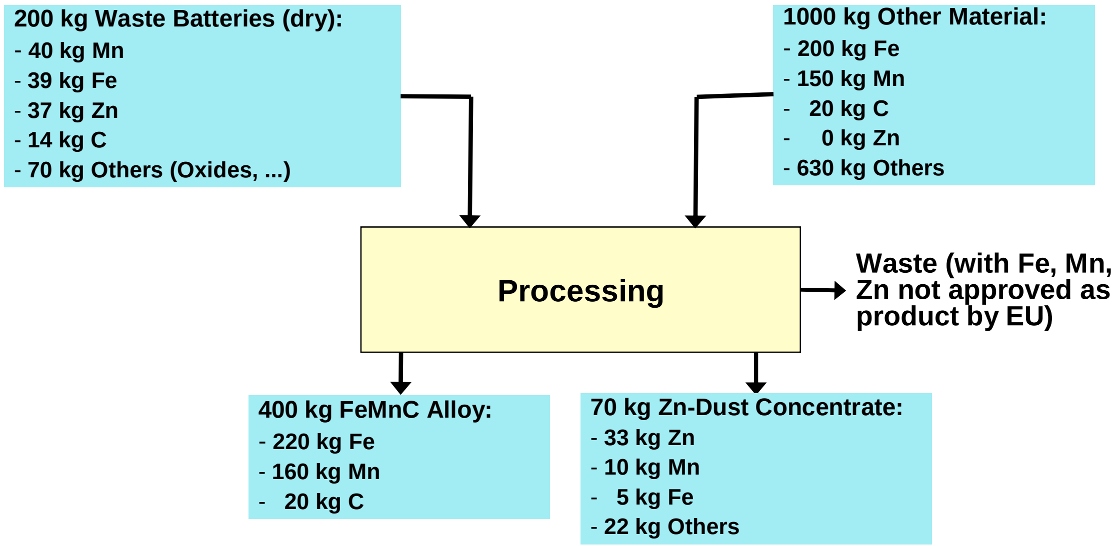
$$RE^{total}: \quad RE_{battery} = RE^X_{battery} + RE^Y_{battery} + \ldots$$

Figure 7: Calculation of recycling efficiency for open loop technology (add to processes)

4.4 Calculation Example

A calculation example for the recycling efficiency of a fictive battery recycling process is shown in Figure 8. The inputs are supposed to be 200 kg waste batteries (dry weight) and 1000 kg other materials. Both fractions contain iron, manganese, zinc and carbon amongst others. Hence the K factors of those elements have to be considered.

The approved products of this fictive battery recycling process are 400 kg FeMnC alloy and 70 kg Zn dust concentrate. All elements of the alloy can be taken into account for the calculation of the recycling efficiency including the carbon because it is captured in a product. The Zn dust concentrate contains zinc, manganese and iron amongst others. Those three metals can also be considered for the recycling efficiency, even MnO_2, if this compound is clearly proved for the dust composition. In our example we assume a different oxide structure (like Fe-Mn-Zn-spinell) and such none of the metal oxides are considered as they are no components of a new battery. The iron, manganese and zinc content of the slag cannot be taken into account at all because in this example we suppose the slag to be classified as waste. The total recycling efficiency of this fictive battery recycling process would amount to $RE_{battery}$ = 57 %.



K^i: $$K^{Fe} = \frac{39kg}{239kg} = 0.16; \quad K^{Mn} = 0.21; \quad K^{Zn} = 1; \quad K^{C} 0.41$$

RE^X: $$RE^{Fe}_{battery} = K^{Fe} \cdot \frac{m^{Fe}_{product}}{m_{battery}} = 0.16 \cdot \frac{(220+5)kg}{200kg} = 0.18$$

$$RE^{Mn}_{battery} = 0.18; \quad RE^{Zn}_{battery} = 0.17; \quad RE^{C}_{battery} = 0.04$$

RE^{total}: $$RE_{battery} = RE^{Fe}_{battery} + RE^{Mn}_{battery} + \ldots = 0.57$$

Figure 8: Calculation example for the recycling efficiency of a fictive battery recycling process

Due to the targeted simplification not all aspects and process details can be reflected. The following examples may show some compromises which have to be accepted:

- If MnO_2 is contained in approved slag or waelz oxide products, it counts as MnO_2.
- If slag is classified as product (non-restrictive of application), it counts in all cases for the recycling efficiency.

- If carbon is extracted as product, separately sold and used, i.e. in or out of a battery recycling process as reducing agent or energy source, it can be taken into account.

5. Summary

According to the new EU Battery Directive battery recycling must reach recycling efficiencies of 65 % by average weight for lead-acid batteries, 75 % for nickel-cadmium and 50 % for other battery types. But up to now no agreement for the calculation method exists. IME Process Technology and Metal Recycling, chair and department of RWTH Aachen University finalized a study aiming on the development of a simplified methodology. In this paper a procedure is proposed, which applies for all battery types and all recycling processes. The outcome of the work can be summarised by the following statements:

- The proposed calculation method is based on relevant elements and compounds which exist in new (virgin) batteries and not on those being present in the scrap. Humidity, plastic shells and electronics or connectors are not considered.
- Battery recycling processes may be based on single or multiple steps plants. Three principle types of recycling processes exist:
    1. closed loop technology (battery to battery)
    2. open loop technology (dedicated battery recycling processes)
    3. open loop technology (add battery scrap to volume of scale processes)
- Materials are products, if not classified as waste with regard to the EU Waste Directive.
- The calculation may end once products are created, which can be used as feedstock for other processes or are individual end-products (independent of market value).

- Each relevant element or compound is considered for the calculation; the total recycling efficiency of the battery recycling process results in the sum of these element/compound specific recycling rates.
- Simplification leads always to compromises, but in this specific case of battery recycling efficiency they have a very small impact on its final value.
- The battery recycling procedure may be split up to various (sub) processes and countries in order to reach economy (average recycling efficiency).

This methodology was developed to stimulate the discussion between politics and battery producers/recyclers as well as their associations in order to find a mutual and long term stable solution for calculating the battery recycling efficiency. The authors do not intend to assess or discriminate existing processes with this proposal. But we have used published data from the recycling sites for validation of the model. Besides some minor compromises to be accepted the method worked well and lead us to the decision of publication. We thank all discussion partners from industry and administration for their fruitful input.

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