

The complexity of defining Recycling Efficiencies in primary battery recycling processes

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Abstract

In July 2006, the European Parliament and the EU Council of Ministers agreed on a compromise to revise the 1991 Directive on portable batteries and accumulators. The New European Directive – published in September 2006 – provides (inter alia) targets for collecting and recycling to be reached by 2016 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 weight for lead-acid batteries, of 75 % for nickel-cadmium and of 50 % on average for all other mixtures of battery types. Due to various reasons the Recycling Efficiency of a process cannot be measured, it has to be calculated. Even though the recycling targets have been fixed, it has not been defined how the Recycling Efficiency is calculated. Critical points to be considered when proposing a calculation method are for example the definition of important and negligible battery components, system boundaries and assigning material values to the input and output materials. Latter ensure the transferability of the method to various recycling processes or even battery types. Furthermore the interpretation of "recycling (product)" has to be taken into account.

In order to provide a strong basis for a constructive suggestion for a calculation method, existing processes that are applied to primary battery recycling in Europe are exemplarily considered. The focus will be on important features such as

- input material-mixes (leading to different values for the Recycling Potential (RP)),
- process modules and process routes of the different recycling systems (leading to suitable material flow calculations and defined system scopes),
- products of the processes (leading to a sufficient comparability of the chosen systems),



- boundary conditions of the calculations

(a. o. limiting the necessary calculation effort for the Recycling Efficiency (RE)).

Based on these calculations different values for theoretically possible Recycling Potentials (RP) and technically feasible Recycling Efficiencies (RE) are indicated and compared to the currently discussed objectives for corresponding Recycling Efficiencies.

1 Introduction

World-wide consumption of primary batteries constantly rises for over 10 years. Presently primary batteries (zinc-carbon and alkaline-manganese) amount the largest percentage of altogether disposed battery systems in Europe and/or Germany (EU: 90 % approx. 150.000 t; Germany: 80 % approx. 30.000 t) [1]. Due to the short life cycle of these battery systems already now - and in the future in ever stronger measure - environmental-relevant material flows have to be controlled and recycled in a suitable way. The associated acute optimisation demand of battery recycling becomes clear by the current European environmental legislation at present:

In the "New EU Directive on portable batteries and accumulators" recycling has for the first time been concretised in data. Apart from the requirement, batteries and accumulators strengthens to collect (defined *Collection Rates*) and to intensify material recycling, in article 19 minimum treatment quotas are defined (*Recycling Efficiencies*). Referring to primary battery types or mixtures on the one hand the Recycling Efficiency of at least 50 % shall be achieved no later than 5 years after entry into force (2011) and on the other hand detailed rules for calculation of the Recycling Efficiency should be added no later than 3.5 years after entry into force (Comitology mid 2010).

2 Characterisation and Recycling Potential (RP) of primary batteries

Against the background of an efficient and thus optimised material recycling, an exact characterisation of the regarded input - as individual material group and/or as representative material mixture - becomes increasingly important.

Depending upon production type and kind of cell, the chemical composition of primary battery systems certain ranges, shown in table 2-1. The main components are manganese dioxide, zinc, iron and electrolytes. Together these constituents build up ca. 80 % (Zn-C) respectively 90 % (Alkaline) of the batteries weight. The content of valuable metals (Mn, Zn, Fe) typically amounts to ca. 55 % (Zn-C) respectively up to 65 % (Alkaline).

Table 2-1: Components of unused primary batteries (data based on [1]-[3])



component	concentration range [%]		
_	zinc-carbon (typical)	Alkaline (typical)	
Mn (as MnO ₂)	14 – 21 (17)	11 – 24 (20)	
Zn (mostly as Zn)	17 – 29 (22)	5 – 18 (16)	
Fe	1-25 (14)	15 – 59 (26)	
C	7 – 11 (9)	0-5 (4)	
Sn	< 0.1	n. v.	
Cu	< 0.2	< 3	
Ni	n. v.	< 0.7	
Hg	< 0.04	< 0.8 * ¹	
organics + paper + bitumen	4 – 13 (8)	3 – 11 (6)	
oxygen (oxides)	< 11	< 12	
electrolyte	$14 - 23^{2} (19)$	5 – 16* ³ (13)	
	$\Sigma_{\text{(typical)}}$: (< 100)	$\Sigma_{\text{(typical)}}$: (< 102)	

^{*1:} prismatical and cylindrical cells: 0 % Hg; button cells: 0,6 - 0.8 % Hg / *2: NH₄Cl, ZnCl₂ / *3: KOH, ZnO

Spent primary batteries enter the metallurgical recycling cycle usually pre-sorted as so-called "market mixtures" (at present approx. 60 % Alkaline and 40 % Zn-C). Different compositions published of such raw material mixtures are summarized in table 2-2.

Table 2-2: Components of typical raw material mixtures ("market mixtures") from primary battery scrap (data based on [1]-[4])

component	average values [%] calculated to given data for			
	unused batteries	"market mixtures"		
Mn (as MnO _X)	18.8	19.4		
Zn (as Zn, ZnCl ₂ , ZnO)	18.4	18.4		
Fe (as Fe, FeO)	21.2	18.9		
С	6.0	7.2		
$SiO_2 + CaO$	n. v.	2.1		
Cu + Sn + Ni + Pb	< 2	< 1.5		
organics + paper + bitumen	6.8	6.5		
oxygen (oxides)	< 12.2	13.0		
moisture (electrolyte)	15.4	12.3		
	$\Sigma < 101$	$\Sigma < 99$		

In the case of primary batteries the content of the valuable main metals (Mn, Fe, Zn) may generally be called *theoretical (relevant metal based) Recycling Potential (RP*_{Me}):

$$RP_{Me} \equiv X_{Zn-C} \cdot (\%Mn + \%Zn + \%Fe) + X_{Alk} \cdot (\%Mn + \%Zn + \%Fe)$$
 (1)



The calculations for "market mixtures" lead to RP_{Me} 's between 57 and 58 % of the input battery weight if the entire weight is taken as reference. For pure alkaline systems the calculated RP_{Me} with approx. 62 % definitely outranges that of the pure Zinc-C systems (approx. 53 %). At the current trend - increasing proportion of alkaline batteries in the market mixtures - this will become relevant in the future. Depending on the kind of the respective mixture and the selected reference basis of the battery weight, the RP_{Me} even ranges between 53 and 83 % (fig. 2-1). Considering that within technical processes metal losses are unavoidable, difficulties in realizing the EU-required Recycling Efficiency of at least 50 % become easy obvious; this concerns in particular Zn-C batteries.

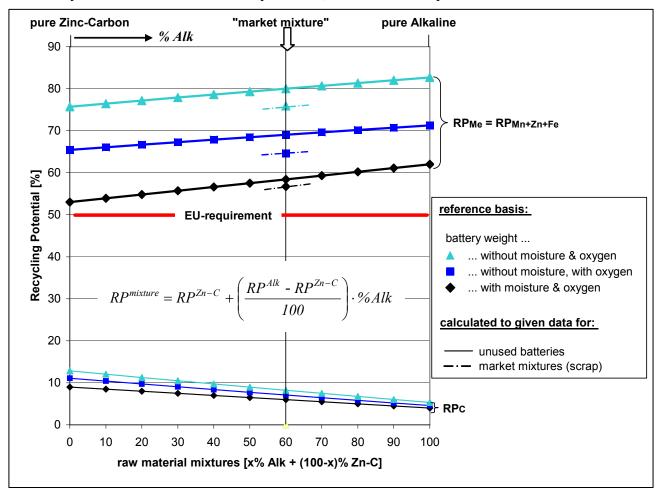


Figure 2-1: Theoretical Recycling Potential (RP) of mixed primary battery scrap

Additionally, the carbon content may be considered (RP_C). According to the recycling process it can be interpreted as efficiency-relevant as well (used as substitute for C-containing reducing agents or as component in FeMn-master alloy). Depending on the kind of the respective mixture and the selected reference basis of the battery weight the RP_C even ranges between 4 and 13 %.

All other components of the primary batteries cannot be recovered by (metallurgical) recycling processes in efficiency-relevant amounts as long as oxides are not treated as products.



3 Recycling alternatives for primary battery scrap

Due to the process engineering more compactly arranged and more effectively working, pyrometal-lurgical recycling processes offer clear cost advantages compared to hydrometallurgical concepts - in particular after elimination of mercury in European primary battery production. Because of high metal contents of manganese, zinc and iron, todays recycling alternatives aim at the reclamation of these valuable metals. In these alternatives however the same input materials partly lead to different output materials (tab. 3-1). For this reason, up to now an objective and well-founded process comparison (e. g. by indication of the Recycling Efficiency) is still lacking.

Table 3-1: Selected recycling procedures for primary battery scrap and associated products

"iron route", e. g.		"zinc route", e. g.		"combined	"combined route", e. g.	
Inmetco; VALDI	DK-Recycling	Mechanical processing/ waelz kiln	Citron S. A.	Batrec (Sumitomo)	IME-concept (Direct SAF)	
	pla	nt capacity (treatn	nent of battery sci	rap)		
commercial (>10.000 t/y)	commercial (<5.000 t/y)	commercial (<5.000 t/y)	commercial (<5.000 t/y)	semi comm. (<5.000 t/y)	pilot plant	
		main	units			
partly shredder → partly pelletising → rotary hearth furnace (only Inmetco) → electric arc furnace	partly sintering → mini shaft furnace	shredder- magnetic sepa- ration → partly pelletising → waelz kiln	rotary hearth furnace	shredder → pyrolysis → induction furnace	partly shred- der-magnetic separation → partly pyro- lysis → electric arc furnace	
	,	standardised prod	lucts (Mn, Zn, Fe	·)		
Ferromangane- se [Fe/Mn>1] (Inmetco); Manganese Steel [Fe/Mn>1] (VALDI)	Pig Iron [Fe/Mn>1]	none	none	Ferromangane- se [Fe/Mn>1], Zinc	Ferromangane- se [Fe/Mn<1], Zinc	
		by-products ((Mn, Zn, Fe)			
Slag	Slag	Waelz Slag	none	Slag	Slag	
		intermediate prod	ucts (Mn, Zn, Fe,)		
Zn-Oxide	"Zn-concentrate"	Fe-fraction, Waelz Oxide	Steel scrap, Zn-Oxide, "Mn- concentrate"	partly Zn-Oxide	partly Fe-fraction	



4 Calculation of Recycling Efficiencies (RE)

The Recycling Efficiency (RE) represents the result of material flow calculations and thus from mass balances whose system boundaries must be carefully defined. It cannot be seen only against the background of single process units and their optimisation. As an evaluation basis it must enable an objective comparison of different recycling technologies, too. A strong basis for a computation methodology of the RE rather implies a sufficient *comparability of the balancing systems concerned*. This should ensure, that within the recycling processes similar by-products are produced from comparable raw materials. To limit the calculation effort of the required system expansion, the efficiency-relevant elements and material flows must be identified and selected.

4.1 System scope and system description

One possible solution is to extend the system boundary so far, that the efficiency-relevant metals (Mn, Zn, Fe) can be followed to a level at which they represent a *replaceable and marketable, standardised metal product* (Crude Steel, Ferromanganese, High Manganese Steel, SHG Zinc) with the corresponding metallic and non-metallic by-products (other metals, metal-containing residues and concentrates, slag). In this case the system scope of commercial recycling alternatives for primary battery scrap should include the production of steel and zinc. Depending on the economic/ecological objectives of the recycling way ("iron route", "zinc route"), the necessary step-by-step modules can be linked differently.

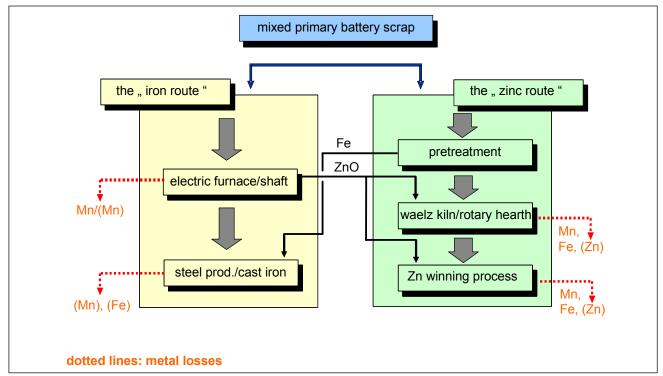


Figure 4-1: System scope of todays commercial recycling alternatives for primary battery scrap For primary battery scrap the definition of the RE may be given by:



$$RE = \frac{m_{s tan dardised products}^{efficiency-relevant elements}}{m_{reference basis of battery weight}^{efficiency-relevant elements}} \cdot 100 \% \equiv \frac{m_{s tan dardised products}^{Mn+Zn+Fe(+C)}}{m_{reference basis of battery weight}^{Mn+Zn+Fe(+C)}} \cdot 100 \%$$
(2)

In order to determine *technically feasible Recycling Efficiencies* and the *influence of material qualities* as well, a brief description respectively definition of the single process modules is essential. This is particularly important

- when materials are integrated in usual technical processes (mostly in limited amounts)
 - → e. g. magnetic iron fraction into steelwork, non-magnetic zinc fraction into waelz kiln, waelz oxide into Zn-electrowinning,
- when only parts of a process are necessary for the recycling of integrated materials
 - → e. g. waelz oxide or "Zn-concentrate" into Zn-electrowinning.

4.2 Boundary conditions

- 1. The calculations of mass balances are based on available data from industry, literature [5-15] and own investigations. In order to limit the necessary calculation effort, an *element-referred cut-off* criterion of 1 % was defined. Thus for (chemical) contents of ≤1 % the appropriate material flow is not continued to pursue concerning this chemical element. This cut-off criterion was used also for dilution effects of particular elements by material integration in usual technical processes with strongly controlled homogenous input mixes.
- 2. In the following evaluation only the oxygen weight of the input-mix is considered. As details are missing, how much oxygen is recovered as zinc oxide or product slag and how much is lost as waste gas, this effect has been fully neglected (*worse case view*).
- 3. Establishing a *total material balance conclusive in itself*, all mass flows were closed according to the method of the error square minimization. If necessary, data were adapted to ensure a 100 % element yield for the calculated recycling process.
- 4. As exemplary "market mixture" the following composition and reference basis was selected:

- 5. The determination of technically feasible values of the Recycling Efficiency (RE) is based on the *specific recoveries* (η) *of the efficiency-relevant elements* in each process module.
- 6. Due to following reasons the *carbon content of the spent primary batteries* was not considered for the calculation of the RE:



- The specific carbon-recovery in the selected process alternatives cannot be seen as a material based recycling but more as a partial chemical/energetic recovery. Whereas the energetic part is clearly excluded by the expected New Directive, the chemical part is still under discussion as a substitute for carbon containing reduction agents.
- Up to now no information is available on the degree of carbon utilization within the relevant step-by-step modules (rotary hearth furnace, EAF, mini shaft furnace, waelz kiln, induction furnace). This is of particular interest because on the one hand some products (ferromanganese, (waelz) slag) contain remaining carbon; on the other hand using the carbon content of battery scrap within the Direct SAF-process a very slow chemical reaction rate was already experienced at the IME.
- Due to the lack of process data, no carbon balance can be calculated.

4.3 Results of mass balance calculations for the mechanical processing/waelz kiln - process

In Germany the mechanical processing/waelz kiln - process represents a best available technique (BAT) of the "zinc route". This recycling alternative can be divided in step-by-step modules as shown in figure 4-2.

- Module 1: mechanical processing

Pre-sorted Zn-C/Alkaline batteries are firstly shredded in a cutting mill and subsequently magnetically separated into an *iron and a non-magnetic share* rich in zinc. The carbon content of the primary battery scrap is recovered together with the non-magnetic share. Exemplary Redux Dietzenbach (Germany) operates such a completely enclosed equipment.

- Module 2: EAF (steelwork)

The *magnetic iron share* – containing minor (non relevant) contents of zinc – is integrated in common steelworks. Together with iron scrap it is melted to *crude steel* in an electric arc furnace (EAF) with addition of fluxes under reducing conditions. The resulting *slag* is used for road construction. Arising flue dusts rich in zinc and lead (*EAF-dust*) are generally processed in waelz kilns followed by Zn-electrowinning.

- Module 3: waelz kiln

The *non-magnetic share* is integrated in common waelz kilns - the world-wide leading process for the Zn/Pb-enrichment of EAF-dust from steelworks (cf. module 2). To feed this large-scale rotary furnace, a homogenous mix of EAF-dust, various zinc-containing residues, coke and fluxes is prepared mostly in form of pellets with an almost constant composition. The main-product of this process is a filter-dust enriched in volatile metal(-compound)s of zinc



and lead (*waelz oxide*), separated in a downstream offgas system. After leaching the alkaline components in a 3-stage countercurrent wash, the waelz oxide can directly fed into electrolytic zinc plants. The by-product *waelz slag* contains all non-volatile components (e. g. Feand Mn-oxides) as well as the fluxes added. Up to now *waelz slag* is still used for landfill site stabilisation and for road construction.

- module 4: Zn-electrowinning (only partly used)

Most of the *SHG zinc* producing facilities in the EU use the electrowinning process - a multi-stage pyro/hydrometallurgical concept for sulfidic zinc concentrates with an increasing share of se-condary raw materials (e. g. *waelz oxide*). While the concentrates pass all process steps – roasting with sulphuric acid plant, (neutral)leaching, leach purification, electrolysis, melting and casting – the feeding of secondary raw materials usually takes place after the (separate) leaching step. On this account, for the calculation of primary battery recycling, only sections of this multi-stage process are to be considered.

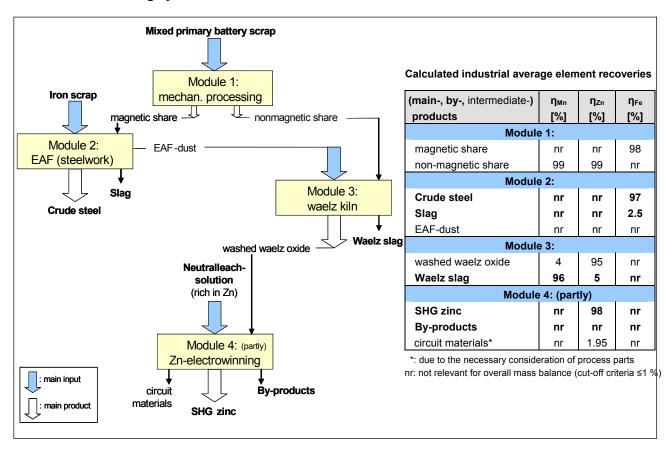


Figure 4-2: Simplified process module flow sheet for the calculated recycling alternative

The calculated *specific element recoveries* (η_{Mn} , η_{Zn} , η_{Fe}) represent industrial (technical feasible) average values; they are summarized in figure 4-2, too. It has to be considered, that the investigated efficiency-relevant elements are not necessarily the main elements of the corresponding single process steps. Summing up all system relevant metal yields of the efficiency-relevant elements it can be



seen, that they are *clearly distributed to the three products - crude steel (Fe)*, waelz slag (Mn), and SHG zinc (Zn) - to a degree of more than 92 % each. The occurring uncertainties of the mass balance result from dilution and distribution effects (remaining flows below the given cut-off criterion, material circuits (external: EAF-dust; internal: various materials from the Zn-electrowinning process)) and rounding errors. They amount to <3 % (Fe), <3 % (Zn) and <5 % (Mn). (fig. 4-3)

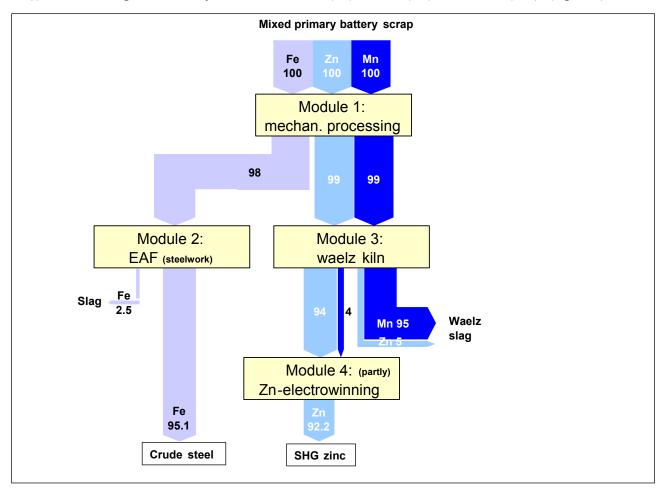


Figure 4-3: Calculated average metal yields of the mechanical processing/waelz kiln - process

The *overall recovery of the batteries metal content is close to 97 %* (if slag counts as recycled material). This clearly underlines the high technological standard of the calculated recycling process. And also the related *Recycling Efficiency* of about 56 % of the total battery weight is even sufficient to fulfil the expected legal threshold. But it has to be assumed that the current practice of using the waelz slag as material for road construction and landfill site stabilisation will be seen more critical in future. Excluding this "by-product" from the calculation of the RE the technically feasible value will be reduced to 37 %. (tab. 4-1)



standard. products	by-products	intermediate products	RE _{Fe} [%]	$rac{\mathbf{RE_{Mn}}}{[\%]}$	RE _{Zn} [%]
Input: "ma	arket mixture" (20.4%	Fe + 18.7% Mn + 18.6% Zn	$\rightarrow RP = 5\%$	7.7 %)	
Crude steel			19.4	-	-
	Slag (steelwork)		0.5	-	-
	Waelz slag		-	17.8	0.9
SHG zinc			-	-	17.1
		(Circuit materials*)	-	-	(<0.05)
		→ RE	19.9	17.8	18.0
<u>R</u> ecycling <u>Efficie</u>	ncy: - with utilization	of slag:		ΣRE_{M}	$_{e} = 55.7 \%$
	- without utilizat	ion of slag:		ΣRE_{Mo}	₂ = 36.5 %

Table 4-1: Calculated Recycling Efficiency (RE) of the mechanical processing/waelz kiln - process

4.4 Comparison of different recycling alternatives

All balances, which are particularly suitable for the calculation of the Recycling Efficiency, are of high sensitivity to the input mix, i. e. its chemical composition. On this account an objective and well-founded process comparison by indication of the Recycling Efficiency is only possible for a *standardised (battery) input mix together with technical feasible metal yields*. Table 4-2 gives an overview of further calculation results – based on identical boundary conditions – for the selected recycling procedures.

It becomes obvious, that all calculated recycling procedures fulfil the legal threshold (RE=50 %) even without carbon-recovery, if the by-product slag counts as recycled material. If not, the processes of the "iron route" and of the "combined route" lead to higher RE values due to the higher metal yield in the metal products. In accordance with the calculations done in this study however the expected legal Recycling Efficiency can not be realised completely by pure metal reclamation (without utilisation of slag). Against this background a strict interpretation of "recycling (product)" becomes more important. For primary battery recycling processes the following scope of interpretation is conceivable:

- recovery of a product, which is equivalent to the original battery raw material
 (e. g.: Zn → zinc-metal in primary batteries),
- reclamation of a product for another industry with an existing standardised market (e. g.: Fe + Mn (+ C) → ferromanganese instead of steel),
- transfer into a by-product, which substitutes natural resources (e. g.: Mn (+ Fe) → slag as mineral additive in road construction),

^{*:} due to the necessary consideration of process parts by the Zn-electrowinning process



• use of a material as reaction agent in metallurgical processes to protect natural resources (e. g.: C (+ Zn) → carbon as chemical reducing agent in Zn-enrichment processes).

Table 4-2: Selected recycling procedures for primary battery scrap and associated products

"iron route", e. g.		"zinc rot	"zinc route", e. g.		route", e. g.
Inmetco; VALDI	DK-Recycling	Mechanical processing/ waelz kiln	Citron S. A.	Batrec (Sumitomo)	IME-concept (Direct SAF)
Recycling	g Efficiency with u	ıtilisation of slag:	RE _{Me} ["market	t mixture": RP _{Me} :	= 57.7 %]
n.a. (Inmetco) ; 54 - 57 %	55 - 57 %	50 - 56 %	50 - 54 %	55 - 57 %	55 - 57,5 %
Recycling I	Efficiency without	t utilisation of sla	g: RE _{Me} ["mark	xet mixture": RP _M	$t_e = 57.7 \%$
n.a. (Inmetco) ; ≤47 %	≤48 %	≤37 %	≤33 %	≤47 %	≤49 %
ave	erage metal yields	in standardised (1	netal) products: η	$\eta_{Me}^{metal\ products} = \eta_{Fe+Mn+Z}^{metal\ pro}$	ducts In
n.a. (Inmetco) ; Fe/Mn, Zn: <85 %	Fe/Mn, Zn: <85 %	Fe, Zn: <65 %	Fe, Mn, Zn: <70 %	Fe/Mn, Zn: <85 %	Fe/Mn, Zn: ≥85 %
		proces	s steps		
<4; <3	<3	<4	<5	<4	<3
		solid wast	e (process)		
++;+	+	++(+)	++	+ (+)	+
		dust emissi	on (process)		
+++;+	++	+++	+++	+ (+)	+

Considering the calculation basis respectively the different reference basis of battery weight, the range and thus the scope for interpreting the results for RE turn out to be rather large, as demonstrated in fig. 4-4. Such a scope of interpretation clearly underlines the imperative for an exact definition as well as a calculation method for the Recycling Efficiency to fulfil the respective legal threshold postulated in the *New European Directive*!



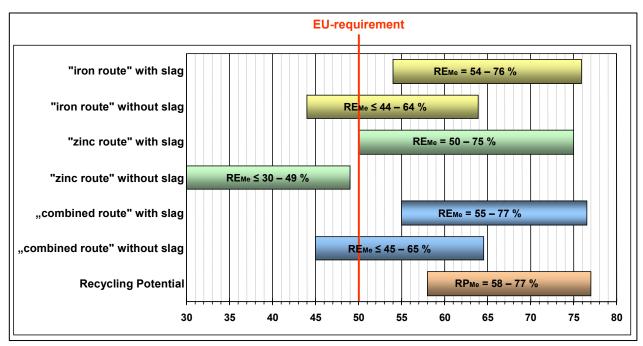


Figure 4-4: Possible interpretation of the calculated Recycling Efficiencies (RE_{Me}) for primary battery recycling processes

5 Summarizing assessment

By introducing regulations for battery recycling in form of defined treatment quotas (e. g. Recycling Efficiency) a common basis is generated, which limits the possibilities of individual interpretation of the particular EU-Member States. The expected legal threshold of at least 50 % by weight for mixtures of all battery types, excepting lead-acid and nickel-cadmium battery scrap, clearly forces recycling activities with optimised material yields. Due to the specific characteristics of primary batteries, their chemical composition and their high amounts of valuable metals (55 - 65 %: Mn + Fe + Zn), the metal industry is particularly challenged to fulfil this limit. Yet, an exact definition as well as a calculation method for determining this Recycling Efficiency is still lacking.

In this study - as a possible input for the current discussion on a qualified calculation method - technically feasible Recycling Efficiencies (RE) of selected recycling procedures were estimated in consideration of the produced material qualities (end-, by- and intermediate products). Summarizing the presented results of the RE-calculations, the following main statements can be given:

- Many alternative reference basis of battery weight theoretically exist to calculate the RE: battery weight with/without moisture & with/without oxygen (oxides).
- If only metals are considered, even theoretically pure Zn-C batteries can't guarantee the legal threshold (reference point: battery weight with moisture & oxygen).



- The interpretation of carbon as efficiency-relevant component as well must be seen more critical, because its partial chemical/energetic recovery and the lacking information on the real degree of carbon utilisation within the relevant technical processes.
- The correct (comparable) definition and calculation of the RE shows a high complexity as downstream modules of other industries than battery recycling industry have to be taken into account.
- An objective and well-founded process comparison by indication of the RE is only possible by regarding a standardised (battery) input mix together with technical feasible metal yields.
- Based on the presented calculation method: independent of the reference basis, all established recycling procedures shall fulfil the legal threshold (RE=50 %) even without carbon-recovery, if the by-product slag counts as recycled material. If not, the procedures of the "iron route" (commercial) and of the "direct route" (still under development) may serve the requirements, as most of the elements are recovered in a metallic stage.

The above statements clarify numerous difficulties to be expected in the near future by technical realisation and legal control of the Recycling Efficiency in the battery industry according to the *New European Directive*. In this context a recommendation – based on the presented calculation method – can be given:

In order to reduce the effort for RE-calculation of each battery recycling company, weighted factors (or rather specific element recoveries) for the necessary downstream modules could be introduced. Such factors should be based on above described mass balance calculations to evaluate own efficiency-relevant material flows and to transfer the in-house RE (only a part of RE!) to an objective, comparable RE of the total recycling procedure, which leads to standardised end-products.

6 Literature

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