

# Contributions of extended batch tests for assessing technical recyclability

## A case study of low-value battery flows

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### Abstract

Good product recyclability is a prerequisite for the transition to a circular economy. However, today's product complexity and diversity in the urban mine result in heterogeneous and variable waste flows affecting process recycling efficiency (RE) and thus product recyclability. For batteries, waste flow composition and subsequent RE are determined by usage behavior, collection, and sorting into chemical battery subsystems. This study aims to demonstrate how extended batch tests (EBTs) can be used as a method to (a) determine battery-specific RE and (b) derive recommendations for assessing and improving the recyclability of batteries. Three EBTs comprising extensive characterization methods were carried out with mixtures of zinc-based (AZ) and lithium-based (LIB) batteries. The results showed that 0.20–0.27 kg/kg of the input mass was lost through flue gas and not recyclable. The metal fraction (0.15–0.19 kg/kg) was easily recyclable, while the mineral fractions of LIBs posed challenges for recycling and recovery (RR) due to the high elemental heterogeneity and pollutants originating from individual battery subsystems. In total, 0.79 kg/kg of AZs was recyclable, whereas 0.52 kg/kg of LIBs and 0.58 kg/kg of AZLIBs (a mixture of both) were recyclable after further treatment. In conclusion, the study demonstrated how the EBT approach can be used to extend recyclability assessment by providing waste flow characteristics for comparison with output quality requirements, enabling assignment of battery-specific RE and identification of poorly recyclable battery subsystems. Thus, the EBT approach can help improve and assess technical recyclability in the future.

### KEYWORDS

batteries, extended batch test, industrial ecology, recyclability, recycling, recycling efficiency

## 1 | INTRODUCTION

In the transition to a sustainable and circular economy (CE), recycling and waste management play a crucial role in closing material cycles and increasing resource efficiency. The European Commission (EC) therefore recently adopted the CE Action Plan (CEAP) promoting CE measures

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for various key product value chains (European Commission, 2020b). Batteries are one example of these key products. Lithium-ion batteries, in particular, have been designated as a key enabler of a low-carbon economy and are notable for their versatile range of uses, rising sales figures (Eurostat, 2019; Kwade & Diekmann, 2018b), and potential fire risk (Kwade & Diekmann, 2018a). One CEAP measure to promote sustainable product designs is the proposal of a new regulatory framework including more systematic durability, recyclability, and recycled content in EU Ecolabel criteria (European Commission, 2020a; European Union, 2020). The design and lifecycle of batteries are therefore becoming the focus of sustainable product policies (European Commission, 2019b) leading to the proposal of an amended battery directive (European Commission, 2020a).

Over the past several years, the design and composition of batteries, especially lithium-ion batteries, have evolved rapidly as a result of product innovations in vehicles as well as in electrical and electronic equipment (EEE) technologies. The increasing demands for battery capacity, efficiency, and cost savings have led to continuous development of new chemical battery subsystems with ever-changing chemical compositions, such as the development from NMC111 to NMC811 (Battery University, 2021; Huisman et al., 2017; Karabelli et al., 2020). This development has resulted in (a) reduction or substitution of cost-intensive battery material such as cobalt and nickel (Huisman et al., 2020) and (b) polymetallic subsystems enhancing battery performance while reducing costs. The resulting product diversity, in combination with varying lifetimes for EEE-specific battery systems (Mählitz et al., 2020) and the hoarding of batteries by end users (Pistoia, 2005), has led to a mixture of battery subsystems in the urban mine with high variability and heterogeneity (Huisman et al., 2017).

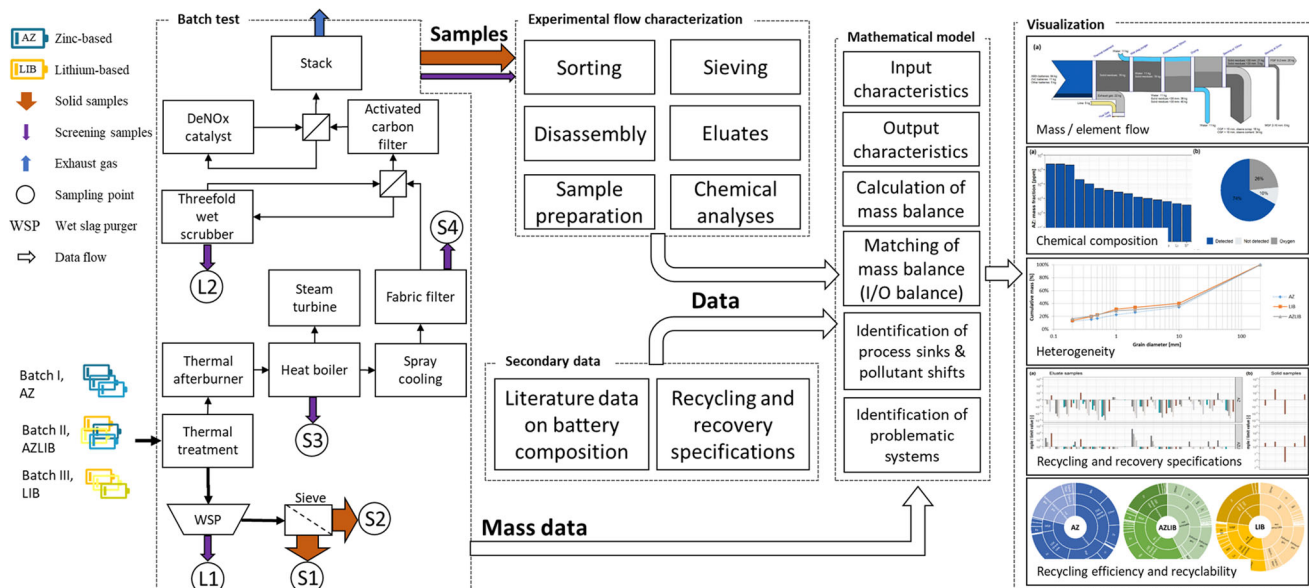
At the end of life (EoL), these battery mixtures require suitable treatment technologies (Mohr, Weil, et al., 2020) to meet recycling efficiency (RE) targets and enable good recyclability (European Commission, 2019a) despite the high material heterogeneity of batteries with critical (Amato et al., 2016) and safety-relevant elements (Huisman et al., 2020). In addition to available treatment capacities, economic and ecological trade-offs, and regulatory requirements (Commission Regulation [EU] No. 493/2012, 2012), the choice of treatment technologies is determined by the composition of battery waste flows, which can be influenced by sorting processes. Given the increasing share of low-value lithium batteries in the main waste flow, the separation of cobalt- and nickel-rich battery subsystems is one possible measure to increase the recovery rate of Co and Ni from the main waste flow in established recycling processes. The result is a waste flow containing various low-value subsystems subjected to alternative and cost-effective recycling processes, such as thermal treatment (Lombardo et al., 2020; Schwich et al., 2021).

In contrast to RE, which is limited to one process and input, recyclability is an indicator for assessing the “ability of a product to be recycled at end of life” (DIN EN 45555, 2019). It comprises a variety of scenarios consisting of setup of processes with individual efficiencies. The recyclability assessment (RA) results are prone to errors, since potential recyclability is based solely on product-related characteristics, such as composition, construction, size as well as (type of) connections between materials and does not include such EoL scenarios and their influence on recycling efficiencies. However, the composition of waste flows in which the product is treated plays a key role in determining process efficiency and, consequently, the recyclability of the waste flow and each product it contains. This influence on recyclability becomes more significant as the constitutional and distributional heterogeneity (Gy, 1995) of the products in the flow increases.

Going beyond potential recyclability requires considering the feasibility of recovery and the respective qualities of anthropogenic resources (UNECE, 2018) to model possible EoL scenarios for RA. Each scenario contains a combination of sorting and treatment scenarios with individual efficiencies that depend on both waste flow composition and treatment technology. The result is an indicator that can be referred to as technical recyclability, which permits a more realistic assessment of product design.

However, including such EoL scenarios in RA is rarely considered and is made challenging by a lack of data. This challenge of collecting harmonized data on waste flows characteristics and process efficiencies to create EoL scenarios can be approached top down (e.g., by modeling; Donati et al., 2020; Wagner et al., 2021) or bottom up (by collecting data on a process level with batch tests). In waste management, batch tests are an established method for internal quality management and assessing process performances (NVMP Association, 2014; Ueberschaar et al., 2017) for waste EEE (WEEE) (Chancerel et al., 2011, 2016; Savi et al., 2013; WEEE Forum, 2013) and EoL vehicles (ELV) (Widmer et al., 2015). Ueberschaar et al. (2017) demonstrated how extending the scope of conventional batch tests can further increase informative value for assessment of element-specific RE by describing the fate of elements, identifying recycling drivers or barriers, assessing output qualities, recommending process optimizations for increased recoveries, and so on.

This study aims to demonstrate the use and design of extended batch tests (EBTs) for characterizing mixed battery waste flows and deriving recommendations to assess and improve product and waste flow recyclability. To achieve these goals, a case study comprising three batch tests with mixtures of low-value (zinc- and lithium-based) battery systems were carried out in a full-scale thermal treatment plant. Comprehensive characterization of input, process, and output in combination with the specification of quality requirements for recycling and recovery serves to determine battery-specific RE and identify battery subsystems that negatively affect RE. The methodology presented using the recycling problem for low-material battery mixtures is applicable to other battery systems and the results can be used to derive recommendations for improved RA as well as for separation and treatment processes of batteries.



**FIGURE 1** Basic flow scheme of the thermal treatment process and description of extensions of the batch test design, based on Ueberschaar et al. (2017)

## 2 | METHODS

### 2.1 | General approach

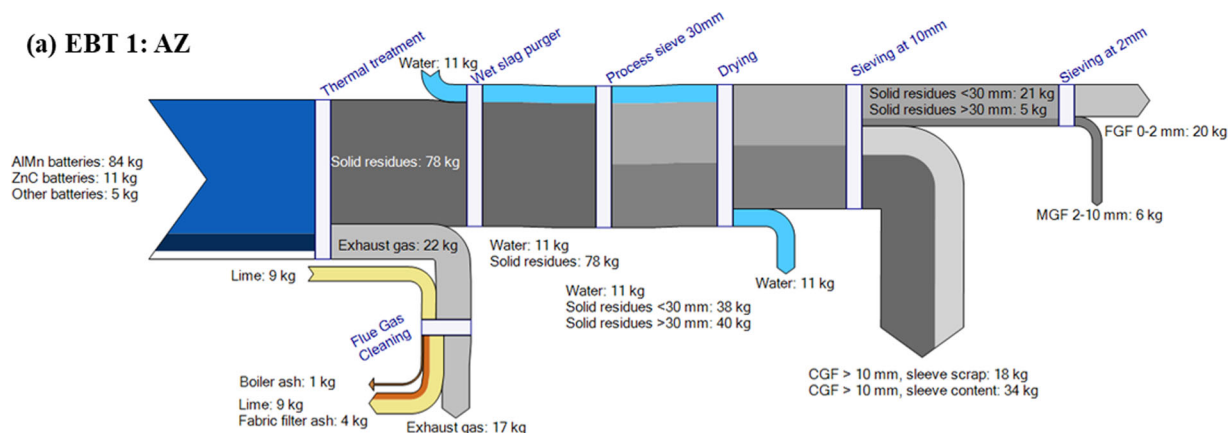
In this study, EBTs were performed based on Ueberschaar et al. (2017) to determine battery-specific RE and assess the recyclability of batteries in mixed waste flows by extending the scope of conventional batch tests by four levels. Firstly, in addition to mass balancing, the EBTs included comprehensive characterization of input and output materials supplemented by screening samples throughout the process to identify process sinks, the fate of targeted elements, and pollutant shifts (see Figure 1). Secondly, the plausibility of the results was checked in terms of mass conservation using mass balancing in an input–output (I/O) balance of selected target elements. Thirdly, the overall batch test RE was calculated by determining the fraction accounting for recycling (Commission Regulation (EU) No. 493/2012, 2012) by comparing output characteristics with quality specifications for possible recycling and recovery options. Fourthly, parameters and elements that would prevent a particular recycling option were identified and studied in terms of their occurrence in specific battery subsystems present in the waste flow. Thus, battery subsystems containing problematic constituents can be prospectively sorted out to prevent reduction of RE and waste flow recyclability and consequently increase the product recyclability of other battery subsystems treated jointly within this waste flow.

### 2.2 | Experimental design of the extended batch tests

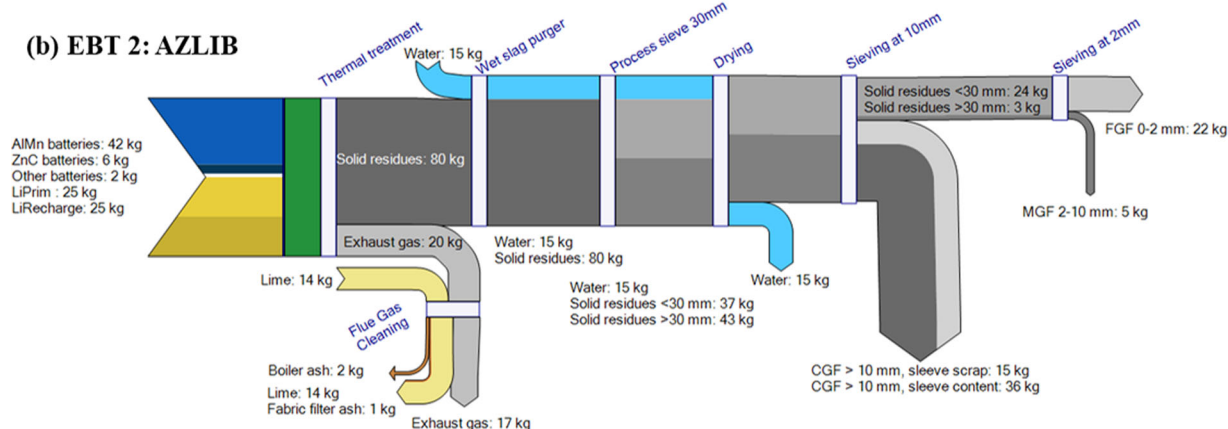
The EBTs were carried out at a full-scale thermal treatment plant for the disposal of ammunition, explosives, and hazardous waste. Input was treated in a ceramic-lined rotary kiln for 30–45 min. at 750–850°C. Flue gas cleaning complies with the emissions limits of the 17th Federal Emission Control Act (Federal Ministry of Justice, 2013) and comprises spray cooling, fabric filter, threefold wet scrubber, activated carbon filter, and SNCR-DeNOx catalyst. The burned-out solid material is discharged via a wet slag purger (WSP) and subsequently sieved on-site with a screen size of 30 mm to separate coarse components (Figure 1, S1) from the fine fraction (Figure 1, S2).

The battery mixtures for the EBTs originated from an industrial sorting plant that separates, among other things, three different battery systems with low intrinsic value. These low-value battery systems are (1) zinc-based batteries (i.e., alkali-manganese “AlMn” and zinc-carbon “ZnC” batteries), (2) primary lithium-based batteries (“LiPrim”), and (3) secondary lithium-based batteries (“LiRecharge”). The three battery systems were provided in 60 L barrels and mixed upon feeding into the treatment plant to simulate different sorting and treatment scenarios (EBT1-3). Under the current battery directive (Directive 2006/66/EC on batteries and accumulators and waste batteries and accumulators and repealing Directive 91/157/EEC, 2006/26.09.2006), all three battery systems belong to the same “other batteries” group, which means that the same RE targets of 50% by mass apply, and the choice of treatment technology is primarily an economic and ecological trade-off. The first and third EBT simulated the EoL scenario with zinc-based and lithium-based batteries treated separately, whereas the second EBT was used to evaluate the possible benefits

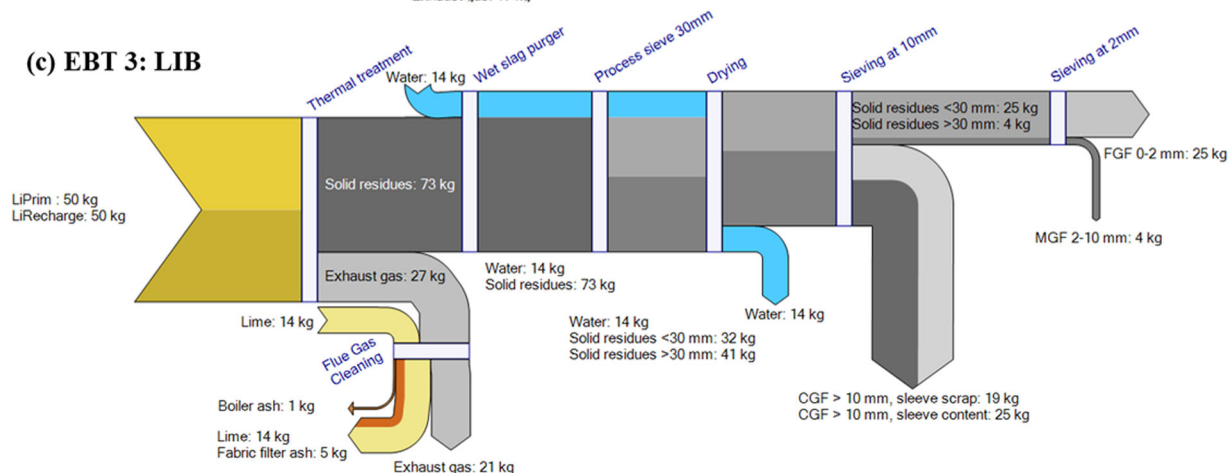
## (a) EBT 1: AZ



## (b) EBT 2: AZLIB



## (c) EBT 3: LIB



**FIGURE 2** Process mass balances for the batches (a) AZ, (b) AZLIB, and (c) LIB. The mass values are normalized to 100 kg input

of treating AZs and LIBs jointly in terms of RE and recyclability. In detail, the three EBT inputs were composed of the following battery systems in percentages by mass:

EBT 1: AZ 100%, in total 9.9 Mg

EBT 2: AZLIB 50% AZ and 50% LIB, in total 10.5 Mg

EBT 3: LIB 50% LiPrim and 50% LiRecharge batteries, in total 10.8 Mg

## 2.3 | Characterization of input material

Since a direct chemical analysis of untreated batteries is challenging in terms of sample preparation and chemical analysis, we chose the method of indirect waste analysis based on product count in combination with a literature review of element mass fraction in batteries. For AZs and LiPrims,

individual samples of 3–4 L batteries were taken upon feeding into the treatment plant over the complete processing time in regular intervals. The total sample mass corresponded to approx. 0.5% of the input quantity supplied. Cylindric, prismatic, coin cells, and pouch cells were sorted by means of labels and imprinted nominal voltages in the subsystems such as AlMn, ZnC, LiMnO<sub>2</sub>/Li(CF)<sub>n</sub> (3 V), LiSOCl<sub>2</sub> (3.6 V), LiSO<sub>2</sub> (3 V), and others. LiRecharge batteries are built into casings, impeding the direct identification of subsystems. Separation of low-value LiRecharge batteries in the industrial sorting plant is based on product-specific use, such as in tools or e-bikes. Thus, three-quarters of these barrels were visually categorized, and the respective product-specific use of subsystems in such devices was researched in the literature (Huisman et al., 2020; Stiftung Gemeinsames Rücknahmesystem Batterien, 2007; Tsiropoulos et al., 2018). The elemental composition of the battery subsystems was determined from the literature (European Portable Battery Association [EPBA], 2007; Stiftung Gemeinsames Rücknahmesystem Batterien, 2007; Würfel & Teipel, 2013) and is provided in Supporting Information S2.

## 2.4 | Characterization of output material

### 2.4.1 | Sampling

As shown in Figure 1, eight samples were taken in total, comprising two samples of the solid output material (S1 and S2 in duplicates), two solid screening samples (S3 and S4), and two liquid screening samples (L1 and L2). The sampling of the flue gas path (boiler ash—S3, fabric filter ash—S4, wet scrubber water “WSW”—L2), and wet slag purger “WSP” (L1) helped to close mass balances and assess pollutant shifts in the process (see Figure 1). The solid output material (S1 and S2) was collected in containers behind the process sieve and sampled according to LAGA PN98 (Länderarbeitsgemeinschaft Abfall [LAGA], 2001). For this purpose, containers were divided into two sectors, from each of which eight individual samples of 5 L were taken and combined to form two laboratory samples (S1a, S1b and S2a, S2b) representing approximately 1% of the total solid output mass. Both laboratory samples were analyzed in parallel for internal quality assurance and consideration of possible uncertainties from sampling. The screening samples were taken toward the end of each test and comprised approximately 1 kg of the solid and approximately 1 L of the liquid samples. The detailed sampling procedure is described in Supporting Information S1.

### 2.4.2 | Physical characterization

After drying (DIN EN 14346, 2007), the solid output samples were sieved in a Haver EML 450 DIGITAL PLUS using sieve trays with the nominal diameter of 0.2, 0.4, 0.5, 1.0, 2.0, and 10.0 mm. The subsequent visual inspection resulted in three different grain size fractions with uniform characteristics, which were merged to examine possible recycling routes and adapt further analyses: fine-grain fraction < 2 mm (FGF), medium-grain fraction 0–10 mm (MGF), and coarse-grain fraction > 10 mm (CGF).

### 2.4.3 | Chemical characterization

Selection of preparation and analysis methods was based on the distinction between metal and mineral fractions. The metal fraction consisting of battery sleeves (i.e., metallic cell casings) and magnetically separated metal occurs in the CGF and MGF, whereas the mineral fraction is present in all three grain size fractions. The metal fraction was analyzed directly by energy-dispersive X-ray fluorescence (ED-XRF), and a larger amount of the battery sleeve sample was analyzed indirectly by melting. After comminution to analytical size, the mineral fraction was analyzed using XRF and wet chemical analysis (for FGF). The elements and parameters studied include Al, As, Ba, C, Ca, Cd, Cl, Co, Cr, Cu, F, Fe, Hg, K, Li, Mo, Mg, Mn, Na, Ni, Pb, S, Sb, Ti, Zn, cyanides, and organic compounds. The detailed method by grain size fraction is described in Supporting Information S1.

## 2.5 | Process and element mass balances

Mass balances were performed for input and output masses as well as for target elements. The input mass was determined at the sorting plant, whereas the output mass is the sum of solid residues, flue gas cleaning residues, and exhaust gas emissions. The solid residues were collected in containers. Boiler ash and filter dust were collected in BigBags and weighed on a truck scale. The mass of added lime was subtracted from the total filter dust mass by counting spray shocks with a defined lime mass. Metal oxides in the FGF were approximated to close mass balances, assuming the main constituents to be present as stable metal oxides (see Supporting Information S1).

Element mass balance was performed for target elements with a high mass fraction (Al, Fe, K, Mn, Zn), hazardous potential (Cd, Pb, Cr), and higher value (Co, Cu, Ni). As shown in Equation (1), the total output mass fraction of the *k*th element ( $\bar{c}_{\text{output}}^k$ ) is the sum of the element mass share



per fraction  $j$  ( $m_j\%$ ) multiplied by the respective average mass fraction of element  $k$  ( $c_j^k$ ):

$$\bar{c}_{\text{output}}^k = \sum_{j=1}^n m_j\% \cdot \bar{c}_j^k \quad (1)$$

To check the plausibility and compliance of input and output, the results of input sorting of battery subsystems were combined with elemental battery contents from the literature (Almeida et al., 2019; Al-Thyabat et al., 2013; EPBA, 2007; Huisman et al., 2020; Mudgal et al., 2011; Tsiropoulos et al., 2018; Würfel & Teipel, 2013).

## 2.6 | Calculation of recycling efficiency and recyclability

Process mass balancing and output characterization are used for calculation of overall process RE, which is legally required for battery recycling processes (Commission Regulation [EU] No. 493/2012, 2012) and described in European Commission (2012). According to Equation (2), RE is the sum of the output flows  $m_{\text{output}}$  accounting for recycling (recyclable content) divided by the input mass  $m_{\text{input}}$ :

$$R_E = \frac{\sum m_{\text{output}}}{m_{\text{input}}} \cdot 100\% \quad (2)$$

In contrast to RE, which describes a treatment step for a specific waste flow and process, recyclability is an indicator that considers the proportion of a product type in different waste flows with different EoL scenarios and efficiencies. However, calculation of recyclability is not legally established for batteries. Following DIN EN 45555 for (W)EEE, recyclability for a specific waste flow and process can be expressed as the quotient of the sum of recycled components of a product divided by the total mass of a product.

## 2.7 | Options for recycling, recovery, and disposal

The focus of recycling and recovery (RR) options was on the solid residues: FGFs, MGFs, and CGFs. No further recycling options were evaluated for flue gas cleaning residues. Identified RR options included (1) metal recycling for pig iron, steel, and copper, (2) recycling as construction materials, and (3) backfilling. In the case that RR is not possible, landfill criteria have been evaluated. RR requirements and limit values were researched in regulations, literature, and through inquiries with metal recyclers that provided product- and process-specific requirements for the respective industry. A general description of the requirements is given in Table 1. Detailed information as well as limit and reference values are provided in Supporting Information S1 and Supporting Information S2.

If a recycling option specifies limit or allocation values, the measurement result is normalized to the limit value by dividing the measurement result by the limit value. If the quotient is greater than 1, the limit value is exceeded. If it is less than 1, the limit value is not reached. For recycling routes in which there are no limit values but instead guidance values for secondary raw materials, the qualities are compared with these requirements.

## 3 | RESULTS

### 3.1 | Input characteristics

The manual sorting of the low-value battery systems AZ, LiPrim, and LiRecharge used to mix the EBT input resulted in the following mass fractions of battery subsystems:

1. AZ: 84% AlMn, 11% ZnCd, 0.1% LiPrim, 0.2% NiCd, 0.7% NiMH, and 4% other batteries
2. LiPrim: 51% lithium-manganese-oxide ( $\text{LiMn}_2\text{O}_4$ ) and lithium graphite fluoride ( $\text{Li}(\text{CF})_n$ ), 39% lithium thionyl chloride ( $\text{LiSOCl}_2$ ), 6.4% lithium sulfur dioxide ( $\text{LiSO}_2$ ), 2.1% LiPrim unknown, 0.8% lithium iron disulfide ( $\text{LiFeS}_2$ ), 0.6% other
3. LiRecharge: 32% power tools (lithium-nickel-manganese-cobalt oxide  $\text{LiNiMnCoO}_2$ : NMC, lithium-manganese oxide— $\text{LiMn}_2\text{O}_4$ : LMO), 32% e-bikes (NMC, LMO, lithium-cobalt oxide  $\text{LiCoO}_2$ : LCO, lithium-iron phosphate  $\text{LiFePO}_4$ : LFP), 20% round cells (NMC, lithium-nickel-cobalt-aluminum oxide  $\text{LiNiCoAlO}_2$ : NCA, lithium titanate  $\text{Li}_4\text{Ti}_5\text{O}_{12}$  or  $\text{Li}_2\text{TiO}_3$ : LTO), 11% flat cells (Li-Polymer), 5% other

**TABLE 1** Material property requirements for selected recycling and recovery options

Recycling and recovery routes		Desired properties	Disturbing properties
Metal recycling	Pig iron "DK process" (DK Recycling und Roheisen GmbH, 2009)	Optimum: Fe contents $\geq 40\%$ by mass, lower Fe contents possible	Solid: Cu, S, Cr, As, Sb, Pb, Co, Mo, Ni, etc., high contents of slag formers ( $\text{SiO}_2$ , CaO, MgO, $\text{Al}_2\text{O}_3$ ) Eluate: –
	Steel (Martens & Goldmann, 2016; Willeke, 1998)	Unalloyed steel: Low content of alloying metals Alloyed steel: Valuable alloying metals	Solid: Cu, S, Sb, As, Sn, Pb, etc. Eluate: –
	Copper (Lossin, 2012; O. Cekel, personal communication, December 22, 2014)	High Cu contents, a favorable ratio of valuable metals and accompanying elements	Solid: High impurity content Eluate: –
Construction materials	Recycling according to LAGA M20 (Joint Working Group of the Federal States on Waste, 2003) and LAGA M19 (Joint Working Group of the Federal States on Waste, 1994)	Low-pollutant, inert material	Solid: TOC, EOX Eluate: Heavy metals (Pb, Cd, Cr, Cu, Ni, Hg, Zn), anions (chloride, sulfate, cyanide), pH value, el. conductivity DOC
Cement industry	Cement industry: Clinker production (Scur, 2013; Verein Deutscher Zementwerke e.V., 2002)  Cement industry: Grinding (InformationsZentrum Beton GmbH, 2014; Verein Deutscher Zementwerke e.V., n.d., 2002)	$\text{SiO}_2$ , $\text{Al}_2\text{O}_3$ , $\text{Fe}_2\text{O}_3$ (CaO) Good bulk material properties (low dust, nonsticky, lumpy), as high homogeneity as possible  Binding effect, improvement of the grain structure in the milling grain range	Solid: Heavy metals and high levels of minor components (MgO, $\text{K}_2\text{O}$ , $\text{SO}_3$ , Cl, etc.) Eluate: –  Solid: Heavy metals Other: Negative impact on the hardening of the cement, the strength and durability of the concrete, the corrosion protection of the reinforcement
Backfilling	Backfilling of waste underground according to Stowage Ordinance (Federal Ministry for the Environment, Nature Conservation & Nuclear Safety, 2002)	Low-pollutant, inert material with low contents of valuable metals	High valuable metal contents Solid: Heavy metals (As, Pb, Cd, Cr, Cu, Ni, Hg, Zn), cyanide (total), organic pollutants, TOC, loss on ignition Eluate: Heavy metals (As, Pb, Cd, Cr, Cu, Ni, Hg, Zn), cyanide (total, easily releasable), organic pollutants
Landfilling	Landfilling according to Landfill Ordinance (Bundesministeriums der Justiz und für Verbraucherschutz sowie des Bundesamts für Justiz, 2009)  Landfill substitute material according to Landfill Ordinance (Bundesministeriums der Justiz und für Verbraucherschutz sowie des Bundesamts für Justiz, 2009)	Compliance with allocation criteria of the respective landfill class  Mineral waste Compliance with allocation criteria of the respective landfill class	Solid: Organic pollutants, loss on ignition, TOC, extractable lipophilic substances Eluate: Heavy metals (As, Ba, Cd, Cr, Cu, Hg, Mo, Ni, Pb, Sb, Se, Zn), anions ( $\text{Cl}$ , $\text{F}$ , $\text{CN}^-$ , $\text{SO}_4^{2-}$ ), total dissolved solids, DOC, phenol index  Solid: Organic pollutants, loss on ignition, TOC, extractable lipophilic substances Eluate: Heavy metals (As, Ba, Cd, Cr, Cu, Hg, Mo, Ni, Pb, Sb, Se, Zn), anions ( $\text{Cl}$ , $\text{F}$ , $\text{CN}^-$ , $\text{SO}_4^{2-}$ ), total dissolved solids, DOC, phenol index

The composition of these fractions was combined with the literature data in the earlier-mentioned mixing ratio for calculation of elemental input composition (see Supporting Information S2).

### 3.2 | Process mass balance

The mass balance results were normalized to 100 kg input masses and are shown in Figure 2a–c. For all batches, 0.20–0.27 kg/kg of the input was discharged via the flue gas cleaning system comprising of exhaust gas (0.17–0.21 kg/kg), boiler ash (0.01–0.02 kg/kg), and filter dust (0.01–0.04 kg/kg). The metal-rich and mineral solid residues left the process via the WSP and account for 0.73–0.80 kg/kg.

After drying and sieve classification of the samples, more homogeneous grain size fractions were obtained. Extrapolated to the normalized input, these grain size fractions account for 0.20–0.25 kg/kg FGF (<2 mm), 0.05–0.06 kg/kg MGF (2–10 mm), and 0.48–0.53 kg/kg CGF (>10 mm) in relation to the normalized plant input (see Supporting Information S2). Comparing the three batches shows similar mass distributions of the output fractions across both the flue gas path and the solid treatment residues. Slight differences are observed for the treatment of LIB.

### 3.3 | Characteristics and recyclability assessment of EBT output

#### 3.3.1 | Coarse-grain fraction (CGF)

After treatment, the CGF contained both completely opened and cracked battery sleeves. The latter still contained their components and fine mineral material, while volatile constituents were released to the gas phase. Subsequent manual sorting and opening of the cracked sleeves resulted in a proportion of 0.15–0.19 kg/kg of pure battery sleeve cases (hereinafter referred to as sleeve scrap) without adhesions and 0.25–0.36 kg/kg of sleeve content. As shown in Figure 3a, the sleeve scrap analysis showed that the AZ is comprised of low-alloyed steel with 0.98 kg/kg Fe, whereas the LIB sleeve scrap contains more alloying elements, such as Ni (0.04 kg/kg), Cr (0.04 kg/kg), and C (0.01 kg/kg).

The sleeve contents of all battery systems contained about 0.50 kg/kg ferromagnetic constituents (Fe, Ni). The ED-XRF screening showed Mn (0.25 kg/kg) and Zn (0.08 kg/kg) as the AZ fraction's main constituents (see Figure 3b). Other constituents were present in low mass fractions. The LIB sleeve content is characterized by higher material diversity with several elements in mass fractions between 0.02 and 0.04 kg/kg (Al, Cu, Fe, Zn, Si, Ni).

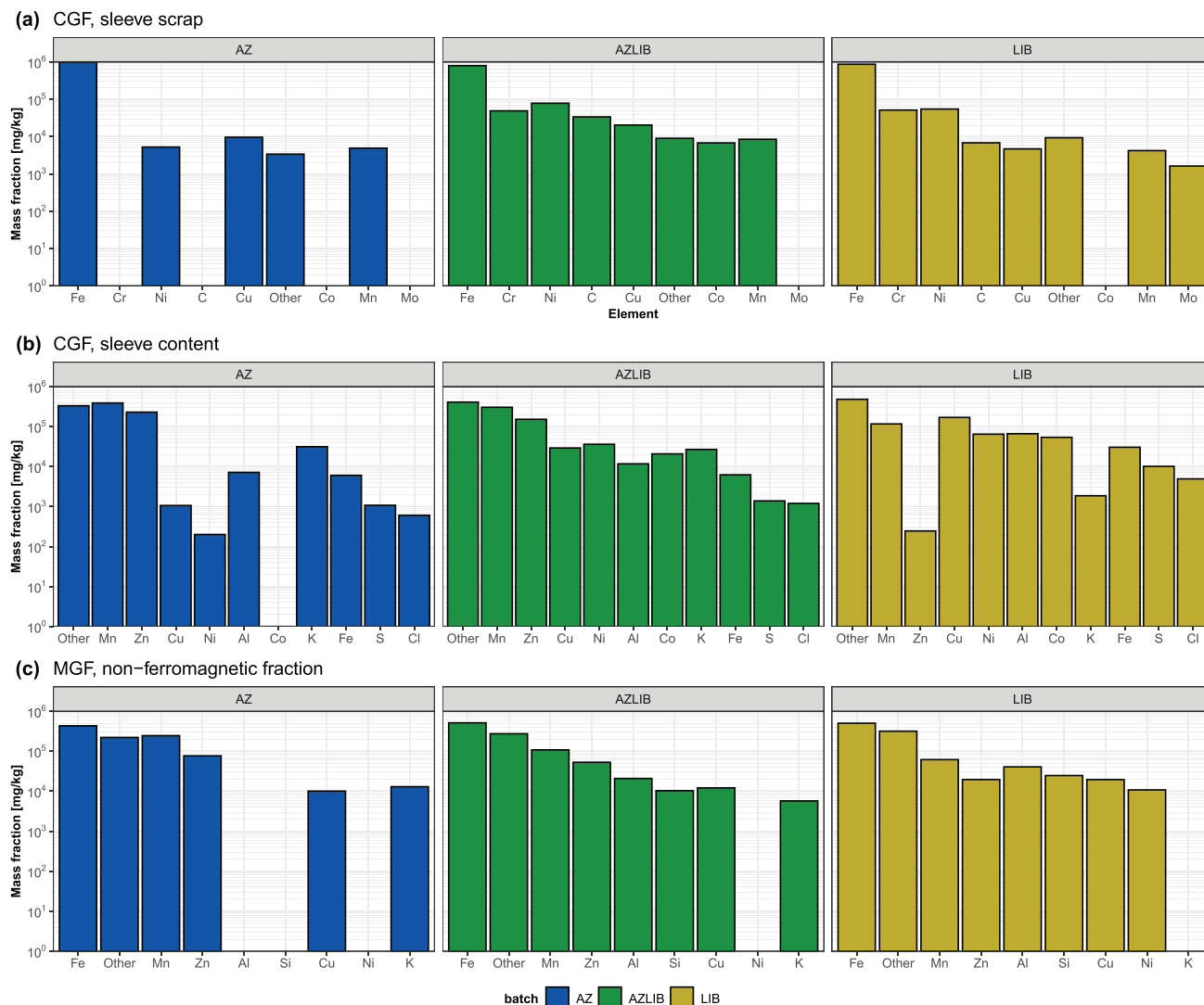
Direct metal recycling of the CGF fraction is only possible considering the impact of pollutants from the more heterogeneous sleeve contents and may require conditioning in line with subsequent use. The investigations showed that the steel qualities in AZs and LIBs are different: low-alloy steels are used in AZ batteries, while higher-alloy steels were identified in LIBs. Joint metal recycling of the sleeve scrap is possible, accepting a downcycling of the LIB steel fraction. Provided that S, Se, Sb, and Cu are within the acceptance range, the LIB sleeve content can be used to produce alloy steel. Due to its high metal content, the LIB sleeve content remaining in the CGF is different from the FGF released by thermal treatment and thus could be of interest for copper recycling with integrated nickel and cobalt recovery. For the AZ coarse fraction, separation of the sleeve scrap from the remaining contents is not necessary. Based on the composition, this fraction could be used to produce a pig iron–manganese alloy (e.g., in the DK process with zinc recovery; DK Recycling und Roheisen GmbH, 2009), provided that it contains no accompanying interfering elements.

#### 3.3.2 | Medium-grain fraction (MGF)

Figure 3c shows the composition of the MGF that differs strongly from the FGF. For all three batches, the magnetic separation resulted in approximately 0.50 kg/kg ferromagnetic metals. The non-ferromagnetic fraction of AZs consists mainly of Mn and Zn (oxidized  $\text{MnO}_2$  and  $\text{ZnO}$ ). Except for slightly higher Cu contents (approximately 0.01 kg/kg), the ED-XRF screening showed no significant impurity contents for the AZ MGF. This Cu content is within the acceptance range, making direct recycling of 0.75 kg/kg of the AZ MGF possible in the DK process or steel processing.

In contrast, the non-ferromagnetic components of the LIB MGF are present in lower mass fractions (0.12 kg/kg Mn, 0.08 kg/kg Al, 0.05 kg/kg Fe, 0.04 kg/kg Cu), which are too low and heterogeneous for recycling in one of the metal routes considered.





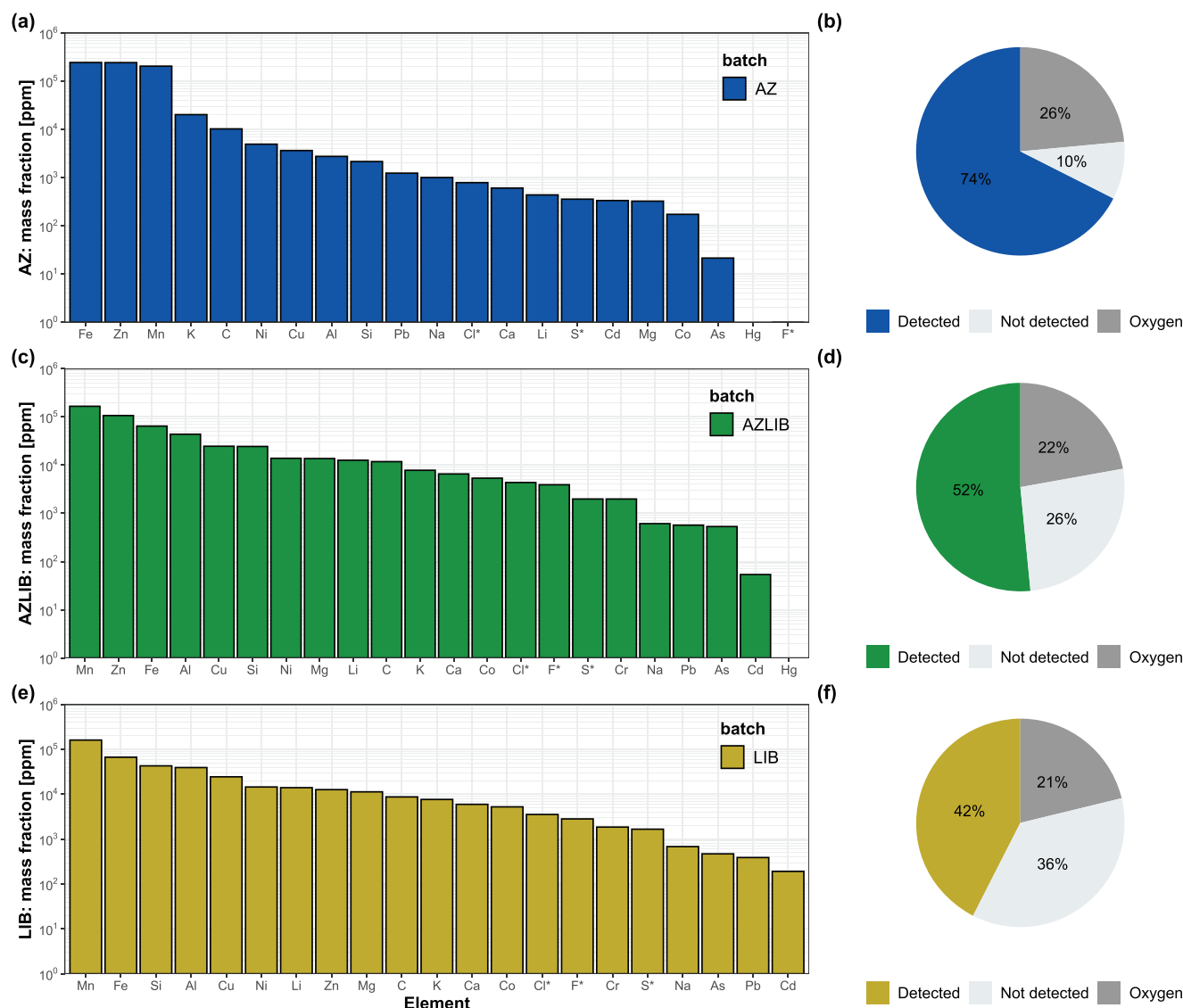
**FIGURE 3** Elemental mass fractions in the coarse-grain fraction (CGF): (a) sleeve scrap, (b) sleeve content as well as the (c) non-ferromagnetic medium-grain fraction (MGF). The underlying data for this figure can be found in Supporting Information S2

### 3.3.3 | Fine-grain fraction (FGF)

The chemical compositions of the burned-out FGF (LOI < 0.01 kg/kg) of AZs and LIBs differ significantly from each other. AZs show higher contents of Fe, Zn, and Mn (>0.10 kg/kg). In contrast, the LIB fine material shows a much higher material diversity. With wet chemical analysis, 0.42 kg/kg (LIBs) to 0.74 kg/kg (AZs) of the FGF material composition could be identified (see Figure 4b,d,f). Assuming that the FGF main components are present as stable metal oxides, calculated oxygen contents of 0.21–0.26 kg/kg are obtained (see Figure 4). The FGF main constituents in the AZ fraction are Fe (0.24 kg/kg), Zn (0.24 kg/kg), and Mn (0.20 kg/kg) with a mass fraction above 0.20 kg/kg TS. The main constituents of LIBs are Mn (0.16 kg/kg), Fe (0.06 kg/kg), Al (0.04 kg/kg), and Cu (0.02 kg/kg), whereas AZLIBs contained mainly Mn (0.17 kg/kg), Zn (0.11 kg/kg), Fe (0.06 kg/kg), Al (0.04 kg/kg), and Cu (0.02 kg/kg). Organic pollutants such as PCBs, PAHs, or CHCs were not detected.

Comparing the FGF characteristics with requirements for metal recycling, construction material recycling, backfilling, and landfilling did not reveal any clear recycling options for all batches. Figure 5 shows the FGF characteristics normalized to the specifications of LAGA M20/M19, landfill classes (LC 0-III), and the Stowage Ordinance for backfilling. The recyclability for metal recycling was estimated via the main components (see Table 1) and validated by individual queries to the respective industry.

For AZs, the high content of Fe, Zn, and Mn (Figure 4a) allows for integrated recycling in pig iron production (DK process), steel recycling, or use as a construction material. However, the limits for halogens and heavy metals in the eluate must be met as a prerequisite. The content of Zn, Ni, and Cu exceeds the specifications (Figure 5b) for backfilling. In the eluate, exceedances of As and Cl are limiting. The DOC content is generally too high for possible landfill. However, given possible alternatives, backfilling or landfilling for AZs is not necessary.



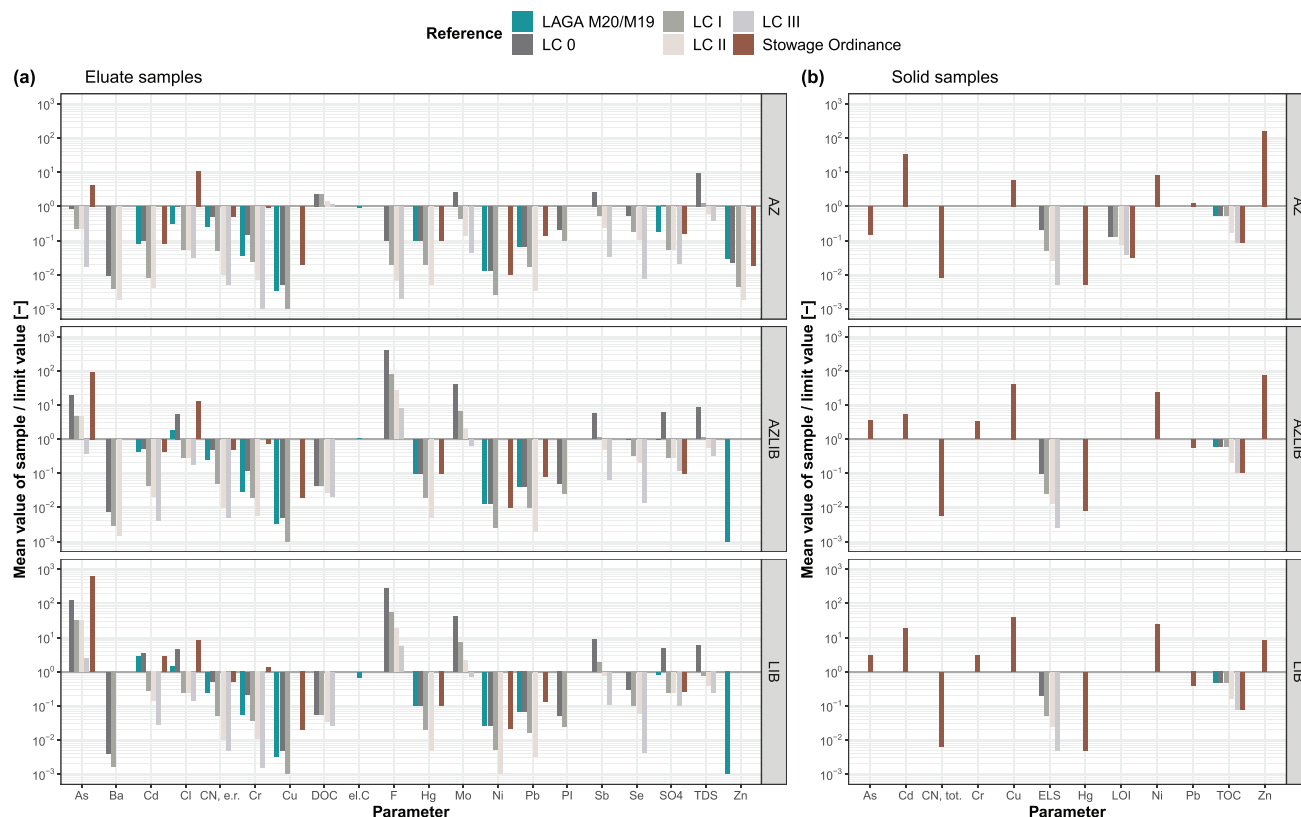
**FIGURE 4** Element mass fraction of the FGF for AZ (a), AZLIB (c), and LIB (e) with the respective mass share of identified elements and oxygen (b, d, f). The underlying data for this figure can be found in Supporting Information S2

The heterogeneous composition of lithium batteries, which contain both valuable and harmful substances in low mass fractions (Figure 4c), impedes RR options. The contents of valuable substances (such as Cu, Ni, Zn, Fe) are too low for metallurgical processes (pig iron, steel, copper; see Supporting Information S1), whereas the allocation values for backfilling are exceeded (Figure 5b). High contents of As, Cd, F, Cl, and Mo in the eluate and As, Cd, and Cr in the solids interfere with all RR options (Figure 5a). High contents of disturbing halogens can be traced back to individual battery systems (e.g., F from  $\text{Li}(\text{CF})_n$  and Cl from  $\text{LiSOCl}_2$ ). However, the As, Cd, and Cl contents are limiting for usage as construction materials. Under certain circumstances, recycling as construction material may be possible after further processing and immobilization or segregation of halogens and heavy metals. Use in the cement industry requires case-by-case examination and further long-term elution tests. The problem of recovering this polymetallic and heterogeneous material is illustrated particularly well by the fact that all landfill class limits are exceeded.

Joint treatment of LIBs and AZs leads to unintended reduction of valuable substances (see Figures 3 and 4) and does not yield compliance with limit values or RR specification. Finding appropriate RR options for AZLIBs is thus approximately as difficult as for LIBs. The complete results can be found in Supporting Information S2.

### 3.4 | Input-output balance

The input-output (I/O) balancing of target elements enables us to draw a conclusion about process sinks and check the plausibility of mass conservation. Elements with more than one mass percent of the total process output (Al, Co, Cr, Cu, Fe, K, Mn, Ni, Zn, Cd, and Pb) were considered. For AZs,



**FIGURE 5** Deviation from the limit value (sample mean/limit) for eluates (a) and solids (b), referencing LAGA M20/M19, landfill classes (LC 0-III), and Stowage Ordinance for backfilling. Easily releasable cyanide (CN, e.r.), dissolved organic carbon (DOC), extractable lipophilic substances (ELS), loss on ignition (LOI), phenol index (PI), total dissolved substances (TDS). The underlying data for this figure can be found in Supporting Information S2

the results in Table 2 show the approximate match (output divided by input O/I) between the element mass in the output and the calculated input composition. For LIBs, slight differences between input and output are observed. Lower Al and Co contents were measured, whereas the Cu, Fe, and Mn contents matched well. Increased Cr and Ni contents in LIBs and AZLIBs can be traced back to the alloyed steel sleeves. The low Al content can be explained, among other things, by the challenges of ED-XRF analysis for light elements in complex matrices (Mählitz et al., 2019). The low Co content in LIBs and AZLIBs is plausible due to the prior separation of cobalt-rich lithium subsystems. In summary, the I/O balancing proves EBT plausibility and shows an approximate match between input and output characteristics. Thus, the EBT approach can serve as a characterization method for complex battery flows to infer their approximate elemental composition.

As part of the revision of the Battery Directive, new measures and requirements will be imposed on lithium-ion batteries, among others, which include, for example, element-specific recycling efficiencies and the verification of heavy metal contents (Cd, Pb) (European Commission, 2020a). The EBT approach can provide this information as demonstrated in Table 2, which shows the element-specific recycling efficiencies of the target elements. The minimum RE ( $RE_{\min}$ ) considers the target element in the recyclable fraction of the output, whereas the maximum RE ( $RE_{\max}$ ) also considers the potentially recyclable fraction. A very low RE is observed for elements in LIB and AZLIB, which are predominantly present in the FGF (e.g., Li).

## 4 | DISCUSSION

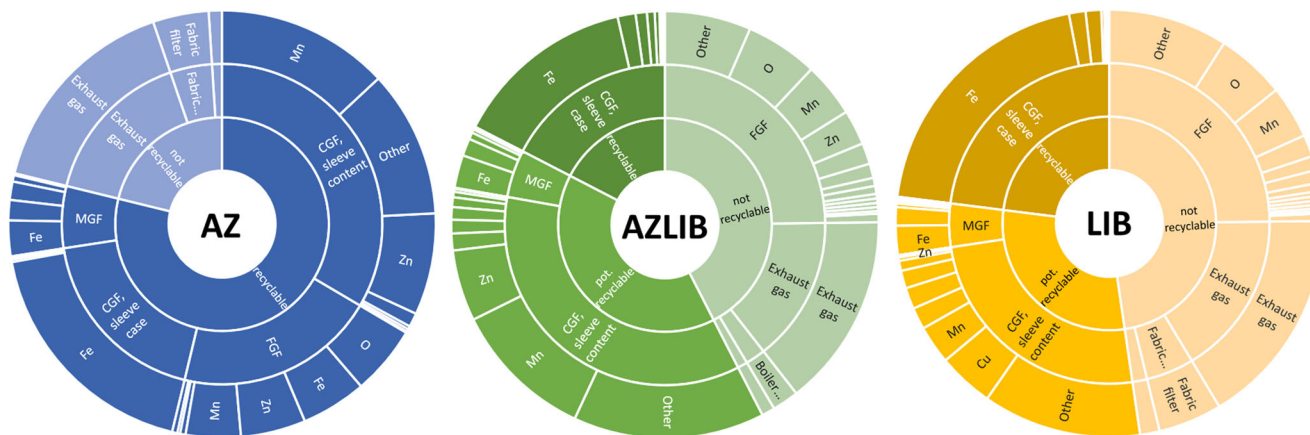
This section discusses the EBT approach's contribution to assessing and improving recyclability and its use in characterizing mixed battery waste flows from the urban mine.

TABLE 2 Mass fraction of selected target elements in input and output (data in mg/kg)

Batch	Element	AZ				AZLIB				LIB				
		Input	Output	O/I	RE <sup>a</sup>	Input	Output	O/I	RE <sub>min</sub>	RE <sub>max</sub>	Input	Output	O/I	RE <sub>min</sub>
Al	3,400	3,000	0.88	1.00	17,400	16,000	0.92	0.00	0.32	27,100	28,000	1.03	0.00	0.65
	100	70	0.70	1.00	80	10	0.13	0.00	0.00	70	50	0.71	0.00	0.00
Co	35	35	1.00	1.00	8,000	9,900	1.24	0.12	0.86	17,300	14,700	0.85	0.00	0.91
Cr	1	0	-	-	4,400	9,000	2.05	0.95	0.95	11,300	12,200	1.08	0.97	0.97
Cu	1,800	3,500	1.94	1.00	21,600	20,500	0.95	0.17	0.70	53,200	50,300	0.95	0.02	0.88
Fe	242,000	262,000	1.08	1.00	179,700	181,000	1.01	0.76	0.91	221,000	246,000	1.11	0.81	0.93
K	14,500	15,000	1.03	1.00	9,900	11,700	1.18	0.00	0.84	2,500	2,400	0.96	0.00	0.19
Li	0	90	-	-	1,500	3100	2.07	0.00	0.00	3,000	3,500	1.17	0.00	0.00
Mn	193,000	189,000	0.98	1.00	162,000	155,500	0.96	0.01	0.73	82,700	72,600	0.88	0.01	0.45
Ni	1,000	2,100	2.10	1.00	23,800	29,800	1.25	0.46	0.88	33,100	32,700	0.99	0.38	0.89
Pb	1,000	250	0.25	1.00	300	140	0.47	0.00	0.00	600	100	0.17	0.00	0.00
Zn	134,000	131,000	0.98	1.00	85,300	82,900	0.97	0.00	0.68	5,000	4,100	0.82	0.00	0.23

Comparison of output and input (O/I) and element-specific recycling efficiencies (RE). RE<sub>min</sub> refers to recyclable content whereas RE<sub>max</sub> includes the potentially recyclable content.

<sup>a</sup>For AZ, RE<sub>min</sub> equals RE<sub>max</sub>.



**FIGURE 6** Proportions of recyclable, nonrecyclable, and potentially recyclable components after further treatment

#### 4.1 | Recycling efficiency of thermal treatment

Thermal treatment allows cost-efficient and safe treatment of low-value batteries with a high explosion risk. The high temperatures crack all sleeves, releasing the electrolyte, rendering the active material inert, and producing an inactive burned-out material comprising metal and mineral fractions. However, in terms of high RE, 0.17–0.22 kg/kg of volatile constituents are lost in the first step via flue gas and are thus non-recyclable (see Figure 6).

While most of the metal-rich CGF and medium-grain fraction (MGF) can be recycled in subsequent metal-recycling processes, the fine-mineral fraction (FGF) of LIBs and AZLIBs consists of heterogeneous material mixtures bearing low contents of valuables and pollutants. The scenario of treating AZs and LIBs jointly did not result in process or output quality improvements with constant or higher RE. Compared to the pure AZ fraction with an overall RE of 79%, AZLIBs result in lower sleeve scrap qualities and higher material diversity of the mineral fraction that lowers RE. For all batches, the proportion of metal scrap is the decisive material fraction relevant for determining total recyclable content and meeting mass-based RE targets. The EBT results showed that the potentially recyclable components could be made available for recycling by subsequent crushing of the closed battery sleeves and adjusting the screen diameter from 10 to 2 mm to separate the heterogeneous fines (FGF). Thus, RE of more than 50% by mass is achievable for lithium-based systems provided that the metal content of battery sleeves does not decrease, which is regarded as critical in view of the low-metal battery pouch cells.

The results indicate that separation and subsequent thermal treatment of low-value batteries can achieve the specified RE targets. Consequently, the legally required RE for the low-value battery mixtures addressed in this study can be achieved with thermal treatment. Given that the mass share of low-value batteries is expected to grow continuously (Huisman et al., 2017), separate treatment of subsystems must be the goal if subsystems are identifiable in sorting. The scenario of separating AZs and LIBs and subsequent recycling (EBT1 and EBT3) enables better recycling and recovery (RR) options due to more homogeneous material characteristics and increases the RE.

#### 4.2 | EBT for recyclability assessment of batteries

The product-centric RA described in DIN EN 45555 (2019) is only applicable if a product is treated and recycled separately. As a result of available collection and treatment infrastructures as well as product identifiability and separability into chemical battery subsystems, however, the recyclability of the product is more dependent on the recyclability of the waste flows in which the products are collected.

Consequently, both easily and poorly recyclable products influence the RE and thus the recyclability of other products in the waste flow. Waste flow characteristics and the selection of recycling processes are thus decisive factors influencing individual product recyclability. These factors must be considered for an improved description of EoL scenarios aiming to include the technical aspects in RA (DIN EN 45555, 2019). However, these data are not yet available on a sufficiently detailed level. EBT can help to fill necessary data gaps in RA here. Batch tests that are legally required to determine RE (Directive 2006/66/EC on batteries and accumulators and waste batteries and accumulators and repealing Directive 91/157/EEC, 2006/26.09.2006; Commission Regulation (EU) No 493/2012, 2012) can be extended by including the product focus on the input and evaluation of potential RR to allow characterization of mixed battery waste flows regarding technical recyclability and allocation of RE to battery subsystems. Further, EBTs can identify battery (sub)systems containing elements such as Cl and F that prevent recycling options by exceeding limit values. Furthermore, the detailed results of the EBT and the comparison with RR options can be used to define end-of-waste criteria (European



Commission, 2018) to create legal certainty and a level playing field as well as remove unnecessary administrative burdens (European Commission, 2021). However, this depth of information requires an equally higher (analytical) effort, as this study shows.

Nevertheless, representative RA results for one battery subsystem require data on the characteristics of all waste flows in which the product is present. Data from different studies on other treatment processes must be collected in a harmonized way to determine the product recyclability independent of a process, providing comparable results that are representative for the corresponding product (Commission Regulation (EU) No 493/2012, 2012). Making results from the EBT approach available, for example, in an urban mine knowledge database (Huisman et al., 2017; Korf et al., 2019; Mähltitz et al., 2020) as a possible extension of the Raw Material Information System (RMIS) (EU Science Hub, 2021), is seen as a long-term goal of improving RA. This knowledge allows the calculation of recyclability in different scenarios and the assessment and definition of targets effectively focusing on the most (environmentally) relevant materials.

For LIBs and AZLIBs, this study showed that separating low-value batteries from the mixed lithium battery waste flow can lower the recyclability of the waste flow. This separation results in lower RE of the considered waste flows, favoring higher RE of the better recyclable and more valuable subsystems of the main flow. Vice versa, without separation of low-value systems from the main flow, RE is decreased in favor of meeting fixed mass-based RE targets. Therefore, we recommended linking efficiency targets to the incoming battery waste flow's recyclability rather than static mass-based targets since maximum material recovery was not always favorable under environmental aspects (Mohr, Peters, et al., 2020). Thus, the safe treatment of currently minor flows of separated low-value battery mixtures becomes feasible and legally compliant. Nevertheless, the results from the EBT approach must be included in future RA to derive both eco-design guidance that includes technical recyclability (DIN EN 45555, 2019) and guidance for recycling processes to cope with challenging waste flows not yet properly recyclable.

### 4.3 | High-quality recycling

High-quality recycling is mentioned in the amended waste framework directive (WFD) (European Commission, 2018), which is an essential component of CE measures and should be prioritized when deciding between recycling options. However, the RE calculation is based on the definition of recycling from the WFD and does not distinguish between high quality and downcycling. At present, there are no standardized methods for evaluating recycling quality. For example, environmental performance (Reuter et al., 2015), environmental relief (Geyer et al., 2016), and the cumulative energy input of recycling processes (Schäfer, 2021) are used to calculate recycling quality. A next step in weighing recycling processes would be development of a systematic and harmonized evaluation method for high-quality recycling based on a solid database of modeling and empirical surveys such as the EBT approach.

In terms of generating pure and defined output qualities, thermal treatment of low-value batteries does not represent high-quality recycling for all scenarios examined here. In particular, the generated mixed mineral fraction of the LIB or AZLIB fraction must be critically considered. After separating the remaining metal components, high-quality recycling of the mineral fraction is hindered by insufficient contents of valuable materials and the presence of disturbing impurities. Thus, the EBT approach can be used to assess high-quality recycling by providing comprehensive output characterization for comparison with detailed recycling specifications. Thermal treatment of the investigated battery mixtures primarily serves the purpose of safe and legally compliant treatment to achieve mass-based RE targets. In the future, backfilling and downcycling of metal alloys will have to be excluded as recycling options in the sense of a CE.

## 5 | CONCLUSION

This study demonstrates how the EBT approach was designed and can be applied to (1) determine RE, (2) approximately infer elemental input composition, and (3) identify problematic battery subsystems that prevent recycling options.

The EBT approach encompassing three scenarios for the thermal treatment of low-value battery systems showed that 0.20–0.27 kg/kg of battery components are lost via flue gas and about 0.15–0.19 kg/kg can be recycled as steel scrap. However, while the mineral fraction of the zinc-based AZ fraction can be recycled (overall RE of 79%), the heterogeneous elemental composition of lithium-based batteries negatively affects the recyclability of the mineral fraction with an overall RE of 0.52–0.58 kg/kg. In particular, individual battery subsystems contribute to the entry of interfering substances such as halogens. Nevertheless, LIBs and AZLIBs can achieve static mass-based RE targets with further processing.

In conclusion, the results from the EBT approach in simulating different collection and sorting scenarios for batteries can serve as the basis for assessing and improving the recyclability of both the treated waste flow and battery subsystems. Harmonized data from sorting and treatment trials are essential for achieving and evaluating CE measures. Furthermore, with appropriate methodological extension of such batch tests, the EoL data generated could serve multiple purposes, such as determining technical recyclability, and should be made available for product design and evaluation of realistically achievable RE.

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## CONFLICT OF INTEREST

The authors declare no conflict of interest.

## DATA AVAILABILITY STATEMENT

The data that supports the findings of this study are available in the supporting information of this article.

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## SUPPORTING INFORMATION

Additional supporting information may be found in the online version of the article at the publisher's website.

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