

Review

Battery Waste Management in Europe: Black Mass Hazardousness and Recycling Strategies in the Light of an Evolving Competitive Regulation

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Abstract: The increasing significance of batteries in the 21st century and the challenges posed by the anticipated surge in end-of-life batteries, particularly within the European context, are examined in this study. Forecasts predict a notable escalation in battery waste, necessitating a focus on the recycling of black mass (BM)—a complex and hazardous byproduct of the battery recycling process. Employing systematic analysis, this research investigates the hazardous nature of BM derived from various battery types. The study underscores the urgent need for definitive legislative classification of BM's hazardous properties (HPs), in accordance with European regulations. This comprehensive examination of BM's HPs contributes significantly to the understanding of BM recycling complexities, proving essential for industry stakeholders and guiding future developments in this field. Additionally, the study explores innovative technologies and strategies that could improve recycling efficiency and reduce associated risks. A pivotal finding of this investigation is the inherently hazardous nature of BM, leading to the recommendation that BM should be classified at a minimum under the “HP3—Flammable” category. This discovery underscores the critical need for stringent management protocols and robust regulatory frameworks to address the burgeoning challenge of battery waste in Europe.



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Keywords: black mass; battery waste; waste classification; battery recycling; hazardous properties; European Regulations; waste management; recycling technologies

1. Introduction

The onset of the 21st century marks a significant escalation in the importance of batteries, a trend anticipated to intensify in the ensuing years. This escalation is not merely a reflection of technological progress but also indicates a fundamental shift in global energy dynamics. With the imminent battery boom, it becomes imperative to comprehend its widespread implications, especially concerning sustainability and resource management. Forecasts for the global battery market suggest a rapid expansion, with projections pointing to a market value exceeding 400 billion dollars and a total size reaching 4.7 TWh by 2030 [1]. This growth, predominantly propelled by the mobility sector, not only underscores an economic opportunity but also presents a series of challenges and responsibilities. A critical challenge in this context is the adaptation of the entire supply chain to accommodate this exponential growth. The battery industry, while flourishing, confronts a crucial limitation in the sourcing of raw materials, raising both availability and ethical concerns [2]. Therefore, a shift towards more sustainable, circular strategies is essential. These strategies encompass extending battery life, planning for their second life, and, most importantly, enhancing recycling processes [3]. Projected data for 2030 highlight an expected increase in end-of-life batteries across Europe, estimated to be around 264,000t. This figure presents a significant challenge when compared to the current recycling infrastructure, capable of handling

only about 80.000t [4]. A crucial aspect of this challenge is the processing of black mass (BM), a complex powder byproduct, difficult to recycle. It results from initial processing stages such as disassembly and pre-treatment, including mechanical size reduction, thermal processes, and density-based separation. This complexity leads to a critical shortfall in efficient and cost-effective technologies for subsequent recycling steps [5]. The difficulties in recycling BM exacerbate the existing imbalance between the demand for and the capacity of recycling solutions. Consequently, bridging this gap requires a comprehensive approach that goes beyond logistical and financial strategies. It calls for an integrated response that combines political will and scientific efforts, working in synergy to develop a sustainable and efficient battery recycling ecosystem. Considering that BM accounts for 40 to 50% of a battery's total weight [6], projections for 2030 underscore a significant challenge in Europe with the management of an estimated 105.000t to 132.000t of this material. The current practice of exporting BM to countries with advanced recycling facilities, while effective in handling the material, leads to a loss of potential revenue for Europe, as BM contains valuable elements like Cobalt and Lithium [7]. This exportation trend stands in contrast to the goals of the Critical Raw Materials Act [8], which aims to reduce Europe's reliance on imported raw materials. Additionally, the BM, despite its free transborder movement, is potentially hazardous. The composition of BM includes substances that are classified as hazardous under multiple EU regulations, starting from the Classification, Labeling, and Packaging (CLP) Regulation (EU) No. 1272/2008 [9], which adheres to the United Nations' Globally Harmonized System (GHS) for the classification and labeling of chemicals. Additionally, these substances are regulated under REACH (Registration, Evaluation, Authorization, and Restriction of Chemicals) Regulation (EU) No. 1907/2006, which is directly applicable in all EU member states [10]. Furthermore, these substances have been identified as hazardous by producers and importers in their notifications to the European Chemicals Agency (ECHA). It is noteworthy that these sources offer slightly different labeling and classifications, with the notifications typically indicating a more extensive range of hazardous aspects. Classifying BM as hazardous waste could potentially restrict its export beyond European borders, thereby ensuring the retention of valuable metals and non-metals within Europe [11]. This approach aligns with the Battery Regulation (EU) 2023/1542 [12], which aims to foster a sustainable internal market by setting specific targets and threshold [13]. Among these, by 2027, the regulation mandates a 50% recovery rate for lithium, escalating to 80% by 2031. For cobalt, copper, lead, and nickel, the targets are set at 90% recovery by 2027, rising to 95% by 2031 [12]. In addressing these challenges, the European Parliament has introduced an amendment, P9_TA (2023)0325, to the proposal for the European Critical Raw Materials Act Regulation. The amendment specifically adds paragraph 7a to Article 25, calling for the development of dedicated waste codes for lithium-ion batteries and their intermediate waste streams, with a particular focus on BM [14].

The primary research objective focuses on a systematic and comprehensive analysis of BM, as sourced from a diverse range of battery types. This involves an extensive collection of BM characterizations available in the current scientific literature, assessing their hazardousness in line with the criteria outlined in Regulation (EU) No. 1357/2014, which defines waste classifications as hazardous and non-hazardous, based on the hazardous properties of the substances they contain [15]. This research objective aims to address the notable lack of specific studies dedicated to this topic and to provide orientation and methodologies for the interested stakeholders. In the dynamic field of battery recycling, the second research question targets the identification of advanced technologies and solutions specifically addressing hazardous elements highlighted in the initial research findings. This inquiry is crucial for guiding investments towards comprehensive solutions, not just interim measures. While the industry eagerly anticipates the development of batteries that are efficient, cost-effective, high-performing, and 100% recyclable with minimal effort, aspiring to reach Technology Readiness Level (TRL) 9, it is also imperative to address current challenges. This balance ensures that in striving for future technological breakthroughs,

the immediate issues in battery recycling are not overlooked but are effectively resolved with targeted and strategic approaches.

2. Materials and Methods

2.1. PRISMA Methodology

In conducting this systematic literature review (SLR), the Preferred Reporting Items for Systematic Reviews and Meta-Analyses (PRISMA) methodology is meticulously adhered to [16]. PRISMA provides a structured framework for gathering, evaluating, and synthesizing scientific evidence, ensuring the transparency and reproducibility of the research process. The initial stage involved a comprehensive literature search. This search was conducted in two main scientific databases: Scopus and Science Direct. The search, conducted on 02 December 2023, used the following string: “black mass” AND (“recycling” OR “management” OR “characterization” OR “Europe” OR “hazardous” OR “regulation” OR “legislation” OR “sustainability” OR “waste” OR “technologies”). This allowed us to collect 1045 potentially useful papers for this SLR. Once collected and stored in files (.res format) containing the basic information of each paper and the related abstracts, they were uploaded to the Rayyan platform to facilitate the Appraisal step [17]. This phase opens with the definition of the eligibility criteria to be used for the preliminary inclusion and exclusion of the papers, which are as follows:

- Inclusion of peer-reviewed research articles, conference papers, and possibly government or industry reports. Exclusion of non-peer-reviewed sources like blog posts, news articles, or opinion pieces.
- Studies specifically focusing on BM in battery recycling, its characterization, management strategies, or related environmental and hazardous aspects.
- Inclusion of research articles published in English.
- Date Range of last 15 years (since 2008).

Through the eligibility criteria, it was possible to proceed to the operational phase of selecting the papers that constitute the basis of the SLR in the Rayyan platform, following the procedure proposed by the PRISMA 2020 flow chart (Figure 1).

At the conclusion of the screening phase, a total of 36 research papers were selected for inclusion in this SLR. Among these, a significant 52.8% were published in the year 2023, followed by 19.4% in 2022, and 13.9% in 2021. To synthesize the findings of this sub-chapter, Table 1 is presented, which serves as a comprehensive repository of the characterizations gathered from the literature, expressed in weight percentage (wt.%). The elements identified in the study are as follows: lithium (Li), cobalt (Co), nickel (Ni), manganese (Mn), aluminum (Al), iron (Fe), carbon (C), cadmium (Cd), potassium (K), titanium (Ti), silicon (Si), calcium (Ca), magnesium (Mg), copper (Cu), zinc (Zn), lead (Pb), phosphorus (P), fluorine (F), sodium (Na), tin (Sn), arsenic (As), and silver (Ag). This table systematically categorizes the content of various substances found in BM, expressed in percentage terms. It also delineates the initial types of batteries from which the BM is derived, classified into distinct categories: lithium-ion batteries (LIBs), nickel–cadmium (Ni–Cd), nickel–metal hydride (Ni–MH), and two separate groups of zinc/manganese batteries—zinc/manganese carbon (Zn/Mn Mn–C) and zinc/manganese (Zn/Mn). The rationale for these distinct categorizations stems from the unique chemical compositions and recycling challenges associated with each battery type:

- Ni–Cd vs. Ni–MH: These batteries are separated due to their differing electrode materials and associated environmental impacts. Ni–Cd batteries contain cadmium, a toxic heavy metal, requiring specific handling and recycling processes. In contrast, Ni–MH batteries, while also based on nickel, replace cadmium with a metal hydride, altering for the composition of the resultant BM.
- Zn/Mn Mn–C vs. Zn/Mn: The division between these Zinc/Manganese battery types is based on their distinct internal chemistries. Zinc/manganese carbon (Mn–C) batteries, traditionally known as alkaline batteries, typically incorporate added carbon, affecting their chemical profile and recycling process. In contrast, standard

Zinc/Manganese batteries, referred to as zinc–carbon batteries, present a different composition, influencing the nature of their BM and are commonly used in household applications.

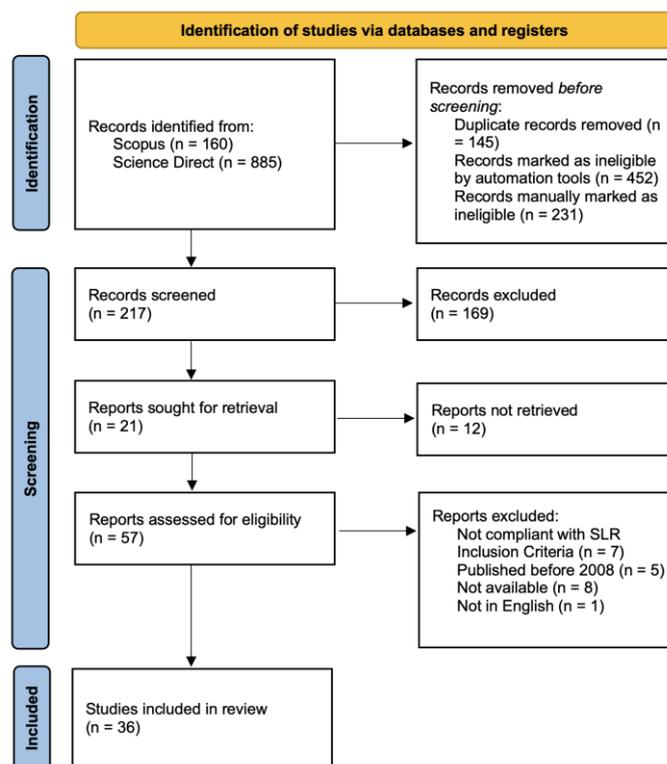


Figure 1. PRISMA 2020 flow diagram illustrating the selection process for research papers.

2.2. Black Mass Hazardousness Classification

The methodology employed to classify the hazardousness of BM in this study is multifaceted and robust, incorporating various regulatory frameworks and a comprehensive database analysis. The initial step involved attributing hazard statement codes (HSCs) to the elements detected in BM. These codes, as defined by the CLP Regulation, REACH, and company notifications, identify specific types of hazards. For instance, lithium is assigned the HSC “H260—Water-react. 1” by the CLP, indicating that it releases flammable gases upon contact with water, which may ignite spontaneously. The HSCs associated with the various elements present in BM are comprehensively detailed in Table 2. In addressing the complexity of BM, which contains elements in a variety of compounds and mixtures, the classification process focused on the most fundamental compound form of each element. This methodology was adopted to ensure a precise assessment of the potential hazards, recognizing that the inherent risk of each element can significantly vary depending on its chemical state within the BM.

Table 1. Characterization of black mass (BM) samples detailed in the scientific literature, expressed as percentages of constituent elements (wt.%).

(wt.%)	1	2	3	4	5	6	7	8	9	10
Type	Mixed LIBs	Mixed LIBs	Mixed LIBs	Mixed LIBs	Mixed LIBs	Mixed LIBs	Mixed LIBs	Mixed LIBs	Mixed LIBs	Mixed LIBs
Li	3.4	9.725 ± 0.055	4.39 ± 0.14	4.302 ± 0.03	3.9	2.6	3.21	3.9 ± 0.1	4.95 ± 1.35	2.95 ± 0.25
Co	17.9 ± 0.1	7.29 ± 0.02	8.34 ± 0.01	4.11 ± 0.029	17.5	5.4	5.3	31.85 ± 0.45	12.65 ± 4.65	24 ± 2
Ni	4.54 ± 0.16	21.395 ± 0.005	16.485 ± 0.085	12.008 ± 0.084	5.1	12.5	14.6		11.4 ± 4.1	11
Mn	5.58 ± 0.08	7.37 ± 0.13	9.39 ± 0.07	6.076 ± 0.042	3	10.9	5.09		11.35 ± 3.75	0.725 ± 0.185
Al	5.405 ± 0.05	7.87 ± 0.11	1.185 ± 0.075	32.815 ± 0.09	1.6	4.8	2.02	0.65 ± 0.25	0.1 ± 0.1	
Fe	1.615 ± 0.185	0.235 ± 0.145		>0	1.6	2.3				0.326 ± 0.234
C				9.17 ± 1.75	36	42.1	39.9	39.6 ± 4.2	28 ± 15.2	
Cd				>0						
K				>0						1.25 ± 0.15
Ti		3.91 ± 0.09		0.15 ± 0.03						0.91
Si	2.115 ± 0.085			>0				1.6	1.15 ± 0.15	
Ca	0.105 ± 0.045			>0						
Mg	0.08 ± 0.04			0.07 ± 0.02						
Cu	3.95 ± 0.05	2.135 ± 0.155	2.11 ± 0.04	5.067 ± 1.403	3.9	3.1	1.83	0.5 ± 0.1	1.2 ± 0.4	6.95 ± 5.05
Zn				0.894 ± 0.045						0.5 ± 0.41
Pb										
P							0.55	0.45 ± 0.05	0.5 ± 0.1	
F							2.5	2.35 ± 0.25	5.7 ± 0.8	
Na										0.315 ± 0.015
Sn										0.72 ± 0.28
As										
Ag										
Source	[7]	[7]	[7]	[5]	[18]	17	[19]	[20]	[20]	[21]

Table 1. Cont.

(wt.%)	11	12	13	14	15	16	17	18	19	20
Type	Mixed LIBs	Mixed LIBs	Mixed LIBs	Mixed LIBs	Mixed LIBs	Mixed LIBs	Mixed LIBs	Mixed LIBs	Mixed LIBs	Mixed LIBs
Li	2.24 ± 0.02	6.96	3.4	3.69	3.9	1.81	1.65	4	3.6	4 ± 0.4
Co	22 ± 0.14		5.8	11.7	24.6	17.1	0.17	32.3	8	26 ± 2.6
Ni	2.71 ± 0.04	0.01	22.2	11.5	3.9	2.22	0.14		7.3	7.3 ± 0.7
Mn	0.75	0.88	7.3	8.91	2.9	4.54	0.1		7.6	3.2 ± 0.3
Al	3.88 ± 0.05	16.4	5.06	2.1	1.6	1.9	3.12	0.9	0.2	1.4 ± 0.1
Fe	6.51 ± 0.12	0.2	0.54		2.5		10.72			
C	20.5 ± 0.27			33.9	33	49.33	37.1	35.4	43.2	
Cd										
K										0.12 ± 0.01
Ti										
Si	0.37 ± 0.07	51.1						1.6	1	
Ca		1.26								0.13 ± 0.01
Mg										
Cu	4.69 ± 0.02	0.11	6.9	0.88	1.1	0.68	0.82	0.6	1.6	2.8 ± 0.3
Zn	0.11									
Pb										
P				0.44			9.64	0.5	0.6	
F				4.1				2.6	6.5	
Na										
Sn										
As										
Ag	0.32 ± 0.01									
Source	[22]	[23]	[24]	[25]	[26]	[27]	[28]	[29]	[29]	[30]

Table 1. Cont.

(wt.%)	21	22	23	25	24	26	27	28	29	30
Type	Mixed LIBs	Ni-Cd	Ni-Cd	Ni-Cd	Ni-MH	Zn/Mn Mn-C	Zn/Mn	Zn/Mn	Zn/Mn	Zn/Mn
Li	3.87		0.03 ± 0.02		0.0515 ± 0.0105					
Co	26.45	1.5 ± 0.1	2.55 ± 2.25	2.4 ± 0.2	7.1 ± 1.2	0.03				
Ni	2.74	37.4 ± 2.1	31 ± 17	44.5 ± 4.5	62.5 ± 1.5	0.28				
Mn	1.67			0.88 ± 0.09	2.45 ± 1.15	43.3	38.7	28.45 ± 4.35	54.7 ± 4.3	30.6
Al	1.64			0.22 ± 0.02	0.675 ± 0.205	0.46	1.06	1.33 ± 0.17	0.79	0.79
Fe	0.61	0.8 ± 0.2	37.08 ± 36.92	3.4 ± 0.5	4.8 ± 1.4	1.42	9.06	8.145 ± 0.055		5.76
C	33	2.4 ± 0.3				8.2		24.6	19	
Cd		27.7 ± 1.6	26 ± 15	20.9 ± 2.1	0.128 ± 0.092	0.01				
K	0.05	2.4 ± 0.2	2.3 ± 1.8	2.8 ± 0.3	2.75 ± 0.35	6.7	0.24	2.735 ± 2.465	0.585 ± 0.085	0.35
Ti					6.35 ± 0.75	0.191				
Si						1.69		5		
Ca	0.03									
Mg	0.09					0.23				
Cu	2.72		0.13		0.15	0.08				
Zn	0.04		0.955 ± 0.645		2.55 ± 0.45	26.88	11.1	11.05 ± 4.05	0.535 ± 0.135	34.8
Pb						0.04				
P	0.45									
F										
Na	0.06		0.515 ± 0.135		0.395 ± 0.155		0.03	0.035 ± 0.005	0.19	0.09
Sn			2.83 ± 1.97		6 ± 1.7					
As					0.41 ± 0.19					
Ag										
Source	[31]	[32]	[21]	[33]	[21]	[34]	[35]	[36]	[36]	[37]

Table 2. Hazard statement codes (HSCs) of the BM elements defined by the CLP Regulation, REACH, and company notifications.

Element	EC/List no.	CAS no.	CLP Regulation HSCs	REACH HSCs	Notifications HSCs
Li	231 102 5	7439 93 2	H314 (Skin Corr. 1B); H260 (Water-react. 1)	H314 (Skin Corr. 1B); H260 (Water-react. 1)	H228; H260; H301; H314; H318; H371; H413
Co	231 158 0	7440 48 4	H317 (Skin Sens. 1); H334 (Resp. Sens. 1) H341 (Muta. 2); H413 (Aquatic Chronic 4)	H302 (Acute Tox. 4); H317 (Skin Sens. 1); H319 (Eye Irrit. 2); H330 (Acute Tox. 1); H334 (Resp. Sens. 1B); H341 (Muta. 2); H360 (Repr. 1B); H373 (STOT RE 2); H400 (Aquatic Acute 1); H410 (Aquatic Chronic 1); H411 (Aquatic Chronic 2); H412 (Aquatic Chronic 3); H413 (Aquatic Chronic 4)	H228; H250; H260; H302; H315; H317; H319; H330; H332; H334; H341; H350; H351; H360; H361; H372; H373; H400; H410; H411; H412; H413
Ni	231 111 4	7440 02 0		H317 (Skin Sens. 1); H334 (Resp Sens. 1); H341 (Muta. 2); H350 (Carc. 1B); H350i (Carc. 1A); H351 (Carc. 2); H360F (Repr. 1B); H372 (STOT RE 1); H411 (Aquatic Chronic 2); H412 (Aquatic Chronic 3)	H228; H250; H317; H334; H341; H350; H350i; H351; H360; H370; H372; H400; H411; H412
Mn	231 105 1	7439 96 5		H411 (Aquatic Chronic 2); H412 (Aquatic Chronic 3)	H228; H260; H302; H312; H315; H319; H332; H335; H340; H341; H360; H361; H370; H372; H373; H411; H412
Al	231 072 3	7429 90 5		H228 (Flam. Sol. 1); H261 (Water-react. 2)	H228; H250; H261; H302; H311; H315; H317; H331; H332; H372; H373; H400; H413
Fe	231 096 4	7439 89 6		H228 (Flam. Sol. 1); H251 (Self-heat. 1)	H228; H251; H261; H302; H315; H319; H335; H370; H371; H372; H373; H400; H410
C	231 153 3	7440 44 0			H226; H228; H251; H252; H300; H302; H315; H319; H335; H373; H412
Cd	231 152 8	7440 43 9		H250 (Pyr. Sol 1); H330 (Acute Tox. 1); H341 (Muta. 2); H350 (Carc. 1B); H361 (Repr. 2); H372 (STOT RE 1); H400 (Aquatic Acute 1); H410 (Aquatic Chronic 1)	H250; H301; H330; H335; H341; H350; H361; H372; H400; H410
K	231 119 8	7440 09 7	H260 (Water-react. 1); H314 (Skin Corr. 1B)	H260 (Water-react. 1); H314 (Skin Corr. 1B); H318 (Eye Dam. 1)	H260; H314; H318; H412
Ti	231 142 3	7440 32 6		H228 (Flam. Sol. 1)	H228; H250; H251; H252; H260; H315; H319; H335
Si	231 130 8	7440 21 3			H228; H315; H319; H335; H373
Ca	231-179-5	7440 70 2	H261 (Water-react. 2)	H261 (Water-react. 2)	H250; H261; H314; H315; H318; H319; H371
Mg	231 104 6	7439 95 4		H228 (Flam. Sol. 1); H250 (Pyr. Sol 1); H252 (Self-heat. 1 and 2); H260 (Water-react. 1); H261 (Water-react. 2)	H228; H250; H251; H252; H260; H261; H315; H318; H335; H413

Table 2. Cont.

Element	EC/List no.	CAS no.	CLP Regulation HSCs	REACH HSCs	Notifications HSCs
Cu	231 159 6	7440 50 8	H411 (Aquatic Chronic 2)	H400 (Aquatic Acute 1); H411 (Aquatic Chronic 2)	H228; H302; H315; H317; H319; H331; H332; H335; H361; H370; H371; H372; H373; H400; H410; H411; H412; H413
Zn	231 175 3	7440 66 6		H410 (Aquatic Chronic 1)	H228; H250; H251; H252; H260; H261; H302; H311; H315; H319; H331; H332; H335; H400; H410; H413
Pb	231 100 4	7439 92 1		H350 (Carc. 1A); H360 (Repr. 1A); H362 (Lact.); H372 (STOT RE 1); H400 (Aquatic Acute 1); H410 (Aquatic Chronic 1); H411 (Aquatic Chronic 2)	H302; H311; H315; H318; H332; H341; H350; H351; H360; H362; H371; H372; H373; H400; H410; H411; H413
P	231 768 7	7723 14 0		H228 (Flam. Sol. 1); H250 (Pyr. Sol 1); H300 (Acute Tox. 1 and 2); H314 (Skin Corr. 1B); H330 (Acute Tox. 2); H400 (Aquatic Acute 1); H412 (Aquatic Chronic 3)	H228; H250; H251; H300; H312; H314; H318; H330; H370; H373; H400; H412
F	231 954 8	7782 41 4	H270 (Ox. Gas 1); H314 (Skin Corr. 1B); H330 (Acute Tox. 2)	H270 (Ox. Gas 1); H280 (Press. Gas Comp.); H314 (Skin Corr. 1B); H318 (Eye Dam. 1); H330 (Acute Tox. 1 and 2)	H270; H280; H310; H314; H318; H330
Na	231 132 9	7440 23 5	H260 (Water-react. 1); H314 (Skin Corr. 1B)	H260 (Water-react. 1); H314 (Skin Corr. 1B)	H260; H314; H318; H370; H412
Sn	231 141 8	7440 31 5			H228; H302; H311; H315; H319; H331; H332; H334; H335; H372; H400; H413
As	231 148 6	7440 38 2	H301 (Acute Tox. 3); H331 (Acute Tox. 3); H400 (Aquatic Acute 1); H410 (Aquatic Chronic 1)	H301 (Acute Tox. 3); H315 (Skin Irrit. 2); H318 (Eye Dam. 1); H331 (Acute Tox. 3); H350 (Carc. 1A); H360D (Repr. 1A); H372 (STOT RE 1); H400 (Aquatic Acute 1); H410 (Aquatic Chronic 1)	H228; H251; H300; H301; H315; H318; H331; H341; H350; H360; H361; H371; H372; H373; H400; H410; H411
Ag	231 131 3	7440 22 4		H360D (Repr. 1A); H400 (Aquatic Acute 1); H410 (Aquatic Chronic 1)	H312; H315; H317; H319; H332; H335; H351; H360; H370; H372; H400; H410; H411

Subsequently, EU Regulation No. 1357/2014 was consulted. This regulation assigns hazard categories to substances based on the presence or exceedance of certain percentages of substances with specific HSCs. Following this, a cross-referencing of the data was conducted. This process involved excluding hazard categories where the substances present in the BM did not contain the required HSCs and analyzing the remaining relevant hazard categories. Table 3 delineates the criteria used to assign hazardous properties (HP) for waste classification, detailing the elements of BM analyzed. For HP2—Oxidizing and HP3—Flammable, the mere presence of specific substances is the determining factor. Other HPs require quantifiable thresholds for each HSC, with some thresholds pertaining to individual substances (Indicated as “Ind.” in the table), such as HP5—Specific Target Organ Toxicity (STOT). Conversely, certain thresholds are cumulative, applying to the total sum of substances within a particular HSC category.

Table 3. EU Regulation No. 1357/2014 hazardousness criteria with corresponding BM elements.

Hazardous Property	HSCs	Element	Condition
HP2—Oxidizing	H270 (Ox. Gas 1)	F	Presence
	H226 (Flam. Liq. 3)	C	Presence
HP3—Flammable	H228 (Flam. Sol. 1-2)	Li, Co, Ni, Mn, Al, Fe, C, Ti, Si, Mg, Cu, Zn, P, Sn, As	Presence
	H250 (Pyr. Sol. 1)	Co, Ni, Al, Cd, Ti, Ca, Mg, Zn, P	Presence
	H251 (Self-heat.1)	Fe, C, Ti, Mg, Zn, P, As	Presence
	H252 (Self-heat. 2)	C, Ti, Mg, Zn	Presence
	H260 (Water-react. 1)	Li, Co, Mn, K, Ti, Mg, Zn, Na	Presence
	H261 (Water-react. 2-3)	Al, Fe, Ca, Mg, Zn	Presence
	HP4—Irritant	H314 (Skin corr. 1A-1B)	Li, K, Ca, P, F, Na
H315 (Skin irrit. 2)		Co, Mn, Al, Fe, C, Ti, Si, Ca, Mg, Cu, Zn, Sn, As, Ag	≥20%—Sum
H318 (Eye dam. 1)		Li, K, Ca, Mg, P, F, Na, As	≥10%—Sum
H319 (Eye irrit. 2)		Co, Mn, Fe, C, Ti, Si, Ca, Cu, Zn, Sn, Ag	≥20%—Sum
HP5—Specific Target Organ Toxicity (STOT)	H370 (STOT SE 1)	Ni, Mn, Fe, Cu, P, Na, Ag	≥1%—Ind.
	H371 (STOT SE 2)	Li, Fe, Ca, Cu, As	≥10%—Ind.
	H335 (STOT SE 3)	Mn, Fe, C, Cd, Ti, Si, Mg, Cu, Zn, Sn, Ag	≥20%—Ind.
	H372 (STOT RE 1)	Co, Ni, Mn, Al, Fe, Cd, Cu, Sn, As, Ag, Pb	≥1%—Ind.
	H373 (STOT RE 2)	Co, Mn, Al, Fe, C, Si, Cu, P, As,	≥10%—Ind.
HP6—Acute Toxicity	H300 (Acute Tox.2 (Oral))	C, P, As	≥25%—Sum
	H301 (Acute Tox.3 (Oral))	Li, Cd, As	≥5%—Sum
	H302 (Acute Tox.2 (Oral))	Co, Mn, Al, Fe, C, Cu, Zn, Sn	≥25%—Sum
	H310 (Acute Tox.1 (Dermal))	F	≥0.25%—Sum
	H311 (Acute Tox.3 (Dermal))	Al, Zn, Sn	≥15%—Sum
	H312 (Acute Tox.4 (Dermal))	Mn, P, Ag	≥55%—Sum
	H330 (Acute Tox.1 (Inhal.))	F	≥0.1%—Sum
	H330 (Acute Tox.2 (Inhal.))	Co, Cd, P, F	≥0.5%—Sum
	H331 (Acute Tox.3 (Inhal.))	Al, Cu, Zn, Sn, As	≥3.5%—Sum
H332 (Acute Tox.4 (Inhal.))	Co, Mn, Al, Cu, Zn, Sn, Ag	≥22.5%—Sum	
HP7—Carcinogenic	H350 (Carc. 1A-1B)	Co, Ni, Cd, As	≥0.1%—Ind.
	H351 (Carc. 2)	Co, Ni, Ag	≥1%—Ind.
HP8—Corrosive	H314 (Skin corr. 1A-1B)	Li, K, Ca, P, F, Na	≥5%—Sum
HP10—Toxic for reproduction	H360 (Repr. 1A-1B)	Co, Ni, Mn, Pb, As, Ag	≥0.3%—Ind.
	H361 (Repr. 2)	Co, Mn, Cd, Cu, As	≥3%—Ind.
HP11—Mutagenic	H340 (Muta. 1A)	Mn	≥0.1%—Ind.
	H341 (Muta. 2)	Co, Ni, Mn, Cd, As	≥1%—Ind.
HP13—Sensitizing	H317 (Skin Sens. 1)	Co, Ni, Al, Cu, Ag	≥10%—Ind.
	H334 (Resp. Sens. 1)	Co, Ni, Sn	≥10%—Ind.

The final stage of the methodology involved a detailed analysis of results from two distinct perspectives. It encompassed an evaluation of individual BM samples, assessing both their minimum and average values, and the construction of theoretical category-specific BM (CSBM). This theoretical construction is based on the original battery types, considering the average values across different BM samples within a category, such as Mixed LIBs, as well as the lowest values observed in these samples. If a substance's minimum value is zero in any category, it is excluded from that CSBM composition (Table 4). Three distinct scenarios were considered for a multifaceted hazard assessment:

- Scenario One: Focused exclusively on the CLP Regulation classifications, this scenario adheres to the GHS criteria and the labeling rules agreed upon by the United Nations.
- Scenario Two: This intermediate scenario broadens the scope by incorporating HSCs from both CLP and REACH classifications, thus expanding the regulatory purview.
- Scenario Three: The most expansive scenario, it compiles all classifications, including those by manufacturers and importers, to reveal the full potential of HPs associated with the BM. This comprehensive view is inclusive of extra-European legislative considerations and provides the most extensive hazard potential profile.

Table 4. Average (Av.) and minimum (Min.) category-specific BM (CSBM) compositions (wt.%).

(wt.%)	LIBs		Ni-Cd		Ni-MH		Zn/Mn Mn-C		Zn/Mn	
	Av.	Min.	Av.	Min.	Av.	Min.	Av.	Min.	Av.	Min.
Li	3.93	1.65	0.01	-	0.05	0.04	-	-	-	-
Co	14.69	-	2.15	0.30	7.10	5.90	0.03	0.03	-	-
Ni	8.05	-	37.63	14.00	62.50	61.00	0.28	0.28	-	-
Mn	4.63	-	0.29	-	2.45	1.30	43.30	43.30	38.11	24.10
Al	4.51	-	0.07	-	0.68	0.47	0.46	0.46	0.99	0.79
Fe	1.29	-	13.76	0.16	4.80	3.40	1.42	1.42	5.74	-
C	22.87	-	0.80	-	-	-	8.20	8.20	10.90	-
Cd	0.00	-	24.87	11.00	0.13	0.04	0.01	0.01	-	-
K	0.07	-	2.50	0.50	2.75	2.40	6.70	6.70	0.98	0.24
Ti	0.24	-	-	-	6.35	5.60	0.19	0.19	-	-
Si	2.81	-	-	-	-	-	1.69	1.69	1.25	-
Ca	0.07	-	-	-	-	-	-	-	-	-
Mg	0.01	-	-	-	-	-	0.23	0.23	-	-
Cu	2.55	0.11	0.04	-	0.15	0.15	0.08	0.08	-	-
Zn	0.07	-	0.32	-	2.55	2.10	26.88	26.88	14.37	0.40
Pb	-	-	-	-	-	-	0.04	0.04	-	-
P	0.63	-	-	-	-	-	-	-	-	-
F	1.13	-	-	-	-	-	-	-	-	-
Na	0.02	-	0.17	-	0.40	0.24	-	-	0.09	0.03
Sn	0.03	-	0.94	-	6.00	4.30	-	-	-	-
As	-	-	-	-	0.41	0.22	-	-	-	-
Ag	0.02	-	-	-	-	-	-	-	-	-

This structured scenario analysis allows for a discerning and layered evaluation of BM hazards, distinguishing the fundamental HP classification from the extended potential risks across varying regulatory frameworks. The approach underpins a thorough appraisal of the BM's intrinsic and potential hazards within the battery recycling milieu.

3. Results

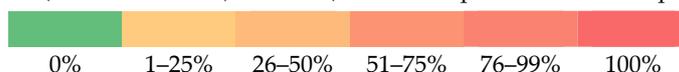
In the results chapter, an intricate analysis was conducted for each HP as defined in the EU Regulation No. 1357/2014. This analysis cross-referenced the conditions of the regulation with the attributed HSCs in the three scenarios outlined in the methodology. The analysis was applied to both individual BM samples and CSBM, assessing their minimum and average values. The findings are summarized in two distinct tables for each scenario, outlining the classification of BM as hazardous waste:

- **Individual Sample Compatibility with HPs:** The first table presents the compatibility of individual samples with each HP, categorized by battery type. Each value represents the percentage of samples in a battery category adhering to a specific HP, ranging from 0% (in green), indicating no sample falls under the category, to 100% (in red), denoting all samples fit the category. These percentages are displayed on a color scale to visually represent the increasing likelihood of BM from a specific battery category falling under the respective HP.
- **CSBM Analysis:** The second table focuses on CSBM, considering the average and minimum values as described in Table 4. It uses a simple “Y” (Yes) in red to indicate classification under a specific HP, or “N” (No) in green when it does not fall under that HP.

3.1. Scenario One: CLP Regulation Classifications

Scenario One is instrumental in understanding which HPs the BM should be classified under, as it solely considers the substance classification provided by the CLP Regulation. A comprehensive overview, synthesized in Tables 5 and 6, reveals that all samples are classifiable under HP3—Flammable due to the presence of Li, K, Na, and Ca, even in the CSBM constructed with the lowest values among the samples. Two other notable categories are HP4—Irritant and HP8—Corrosive. The BM derived from LIBs and nickel-based batteries shows a significant presence of elements with the H314 code (Skin corr. 1A and 1B) exceeding 1%. Specifically, Li in the former manifests a fluctuating presence ranging from 1.65% to 9.73%, whereas K in the latter is observed within a narrower band of 2.3% to 2.8%. It is noteworthy that all LIBs samples that do not fall under HP4—Irritant have a percentage of Li above 5%, classifying them in the HP8—Corrosive category. Furthermore, nearly all the LIBs and nickel-based BM samples are classified as HP7—Carcinogenic due to more than 0.1% presence of Co. Indeed, with the exception of Sample 12, Co is present in quantities ranging from a minimum of 0.17% in Sample 17 to a maximum of 32.3% in Sample 18. These samples are also classified under HP10—Toxic for reproduction and HP11—Mutagenic criteria, requiring the presence of Co to be equal to or exceed 0.3% and 1%, respectively, thus excluding Sample 17 as well. The analysis reveals that BM originating from LIBs is potentially classified under nine distinct HPs, in contrast to zinc-based BMs, which may be categorized under two HPs. This distinction arises from the LIBs BM samples containing adequate amounts of Li, Co, and F. For BMs from Zn/Mn Mn-C and Zn/Mn, the presence of K and Na results in a definitive classification of the former as HP8—Corrosive, while the latter is likely to be classified under HP4—Irritant.

Table 5. Hazardousness classification of Individual BM samples under Scenario One, ranging from 0% (No involvement) to 100% (All BM samples falls under a specific HP).



Scenario One Classification for Individual Samples	LIBs		Ni-Cd		Ni-MH		Zn/Mn Mn-C		Zn/Mn	
	Av.	Min.	Av.	Min.	Av.	Min.	Av.	Min.	Av.	Min.
HP1—Explosive	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
HP2—Oxidising	29%	29%	0%	0%	0%	0%	0%	0%	0%	0%
HP3—Flammable	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%
HP4—Irritant	62%	62%	100%	67%	100%	100%	0%	0%	25%	0%
HP5—Specific Target Organ Toxicity (STOT)	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
HP6—Acute Toxicity	29%	29%	0%	0%	0%	0%	0%	0%	0%	0%
HP7—Carcinogenic	95%	95%	100%	100%	100%	100%	0%	0%	0%	0%
HP8—Corrosive	38%	38%	0%	0%	0%	0%	100%	100%	0%	0%
HP9—Infectious	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
HP10—Toxic for reproduction	90%	90%	100%	100%	100%	100%	0%	0%	0%	0%
HP11—Mutagenic	90%	90%	100%	67%	100%	100%	0%	0%	0%	0%
HP12—Release of an acute toxic gas	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
HP13—Sensitising	57%	52%	0%	0%	0%	0%	0%	0%	0%	0%
HP14 - Ecotoxic	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
HP15—Latently Hazardous Waste	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%

Table 6. Consolidated hazardousness classification for CSBM in Scenario One, denoted by “Y” (Red) for “Yes”, indicating classification under specific HP, and “N” (Green) for “No” involvement.

Scenario One Classification for Category-Specific BM	LIBs		Ni-Cd		Ni-MH		Zn/Mn Mn-C		Zn/Mn	
	Av.	Min.	Av.	Min.	Av.	Min.	Av.	Min.	Av.	Min.
HP1—Explosive	N	N	N	N	N	N	N	N	N	N
HP2—Oxidising	Y	N	N	N	N	N	N	N	N	N
HP3—Flammable	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y
HP4—Irritant	N*	Y	Y	N	Y	Y	N*	N	Y	N
HP5—Specific Target Organ Toxicity (STOT)	N	N	N	N	N	N	N	N	N	N
HP6—Acute Toxicity	Y	N	N	N	N	N	N	N	N	N
HP7—Carcinogenic	Y	N	Y	Y	Y	Y	N	N	N	N
HP8—Corrosive	Y	N	N	N	N	N	Y	Y	N	N
HP9—Infectious	N	N	N	N	N	N	N	N	N	N
HP10—Toxic for reproduction	Y	N	Y	Y	Y	Y	N	N	N	N
HP11—Mutagenic	Y	N	Y	N	Y	Y	N	N	N	N
HP12—Release of an acute toxic gas	N	N	N	N	N	N	N	N	N	N
HP13—Sensitising	Y	N	N	N	N	N	N	N	N	N
HP14—Ecotoxic	N	N	N	N	N	N	N	N	N	N
HP15—Latently Hazardous Waste	N	N	N	N	N	N	N	N	N	N

* The concentration of substances bearing the H314 code exceeds 5%, classifying the BM under HP8.

3.2. Scenario Two: CLP Regulation and REACH Classifications

Scenario Two extends the analysis to encompass REACH classification, marking a significant divergence from Scenario One (Tables 7 and 8). Beyond reinforcing the HPs identified earlier, this scenario introduces additional categories. This results in the attribution of additional HPs to the BM from LIBs and nickel-based batteries. In HP5—STOT, BM is classified due to the presence of Ni and Cd at or above 1%, recognized as H372—STOT RE 1, and/or Co at 10% classified as H373—STOT RE 2 under REACH. For HP6—Acute Toxicity,

Table 10. Consolidated hazardousness classification for CSBM in Scenario Three, denoted by “Y” (Red) for “Yes”, indicating classification under specific HP, and “N” (Green) for “No” involvement.

Scenario Three Classification for Category-Specific BM	LIBs		Ni-Cd		Ni-MH		Zn/Mn	Mn-C	Zn/Mn	
	Av.	Min.	Av.	Min.	Av.	Min.	Av.	Min.	Av.	Min.
HP1—Explosive	N	N	N	N	N	N	N	N	N	N
HP2—Oxidising	Y	N	N	N	N	N	N	N	N	N
HP3—Flammable	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y
HP4—Irritant	Y	Y	Y	N	Y	Y	Y	Y	Y	Y
HP5—Specific Target Organ Toxicity (STOT)	Y	N	Y	Y	Y	Y	Y	Y	Y	Y
HP6—Acute Toxicity	Y	N	Y	Y	Y	Y	Y	Y	Y	Y
HP7—Carcinogenic	Y	N	Y	Y	Y	Y	Y	Y	N	N
HP8—Corrosive	Y	N	N	N	Y	Y	Y	Y	N	N
HP9—Infectious	N	N	N	N	N	N	N	N	N	N
HP10—Toxic for reproduction	Y	N	Y	Y	Y	Y	Y	Y	Y	Y
HP11—Mutagenic	Y	N	Y	Y	Y	Y	Y	Y	Y	Y
HP12—Release of an acute toxic gas	N	N	N	N	N	N	N	N	N	N
HP13—Sensitising	Y	N	Y	Y	Y	Y	N	N	N	N
HP14—Ecotoxic	N	N	N	N	N	N	N	N	N	N
HP15—Latently Hazardous Waste	N	N	N	N	N	N	N	N	N	N

4. Discussion

A pivotal aspect of this analysis is the comparison of CSBM across different scenarios, with a particular focus on Table 10, which contrasts the minimum values in Scenario One and the average values in Scenario Three for CSBM. Scenario One serves as the baseline of our analysis. Here, the classification is conservative, primarily driven by the flammability risk due to the presence of elements like Li, K, Na, and Ca, which directly contribute to HP3—Flammable. In this baseline scenario, other substances such as F play a role in HP4—Irritant and HP8—Corrosive, while Co influences HP7—Carcinogenic; HP10—Toxic for reproduction; and HP11—Mutagenic. In contrast, Scenario Three is recognized as the most precautionary, given that it considers a wider array of properties. By incorporating average values in each CSBM, Scenario Three takes into account the international concern related to BM elements, revealing a broadened hazard profile. The final line of Table 11 contemplates a generic BM classification based on HPs that appear across all CSBMs. This reveals that, within the baseline scenario, HP3—Flammable is the sole HP involved, dictated by the aforementioned elements.

To address the flammability issue associated with BM during the recycling processes, the risk mitigation strategies proposed by [38] can be considered. In this work, the author examines various methods to reduce the fire hazard during the preliminary stages of the recycling process. First, discharging the spent batteries is crucial to prevent sparks or explosions during dismantling. This is achieved mainly through three methods: electrolytic discharge using salt solutions, ohmic discharge using an external circuit, and cryogenic discharge with liquid nitrogen or in a vacuum atmosphere. Each of these methods has specific advantages and disadvantages, such as the use of alkali salts to prevent corrosion in electrolytic discharge, or the limited effectiveness and high costs of cryogenic discharge. Second, for industrial-scale recycling, the method of shredding the batteries in a protected environment is often preferred to reduce the fire risk. Techniques such as the use of water sprays or nitrogen gas, CO₂ atmospheres, or inert gases like argon or nitrogen during shredding have been implemented to prevent the oxidation of lithium and other reactive elements in the batteries. Further studies have indicated additional significant innovative pre-treatment that can be integrated to enhance both the safety and efficiency of the process. The significance of crusher grid size and pre-treatment temperature in the processing of BM is highlighted by Wilke et al. [39]. It has been observed that finer grid sizes and optimized

pre-treatment temperatures can effectively minimize the formation of fine, potentially flammable particles. This approach not only contributes to a reduction in flammability risks but also facilitates the management of particle size distribution, crucial for subsequent processing stages. Punt et al. [40] focus on the method of discharging batteries using a 5% NaCl solution. This technique serves a dual purpose: it mitigates the risk of hazardous reactions inherent in the recycling process and enhances the recovery efficiency of valuable metals. The reduction of reactive and flammable components within the BM through this method renders the material safer for handling and further processing. The concept of mechanical activation, particularly through milling, as a means to influence the reactivity and reducibility of BM is introduced by Babanejad et al. [29]. This process not only advances the energy efficiency of the overall recycling procedure but also impacts the particle size, leading to a more controlled reduction process. The alteration in particle size brought about by mechanical activation plays a pivotal role in managing the flammability risks associated with BM. Moreover, integrating machine learning techniques in the sorting phase can significantly enhance the precision of battery component classification, thereby aiding in the selection of optimal pre-treatment methods and reducing the risk of flammability [41,42]. The preceding sections have provided an in-depth analysis of pretreatment strategies essential for the conditioning of BM from LIBs. These strategies facilitate the separation of materials and enhance the reactivity of the input for recycling processes. Moving forward from pretreatment to the recycling of BM, a detailed synopsis is presented in Table 12, in which a diverse array of BM recycling methodologies, characterized by their distinct operational principles, recovery efficiencies, and inherent limitations. The TRL of each technology were determined through a comprehensive evaluation process. This process included reviewing academic literature and comparing the studies with established TRL definitions outlined by the European Commission [43]. Key factors considered in this evaluation were the development stage, operational data, and technology maturity indicators of each methodology.

Table 11. Range of HPs classification of CSBM and generic BM.

CSBM	Baseline Scenario	Comprehensive Scenario
LIBs	HP3—Flammable; HP4—Irritant;	HP2—Oxidizing; HP3—Flammable; HP4—Irritant; HP5—Specific Target Organ Toxicity (STOT); HP6—Acute Toxicity; HP7—Carcinogenic; HP8—Corrosive; HP10—Toxic for reproduction; HP11—Mutagenic; HP13—Sensitizing
Ni-Cd	HP3—Flammable; HP7—Carcinogenic; HP10—Toxic for reproduction;	HP3—Flammable; HP4—Irritant; HP5—Specific Target Organ Toxicity (STOT); HP6—Acute Toxicity; HP7—Carcinogenic; HP10—Toxic for reproduction; HP11—Mutagenic; HP13—Sensitizing
Ni-MH	HP3—Flammable; HP4—Irritant; HP7—Carcinogenic; HP10—Toxic for reproduction; HP11—Mutagenic	HP3—Flammable; HP4—Irritant; HP5—Specific Target Organ Toxicity (STOT); HP6—Acute Toxicity; HP7—Carcinogenic; HP8—Corrosive; HP10—Toxic for reproduction; HP11—Mutagenic; HP13—Sensitizing
Zn/Mn Mn-C	HP3—Flammable; HP8—Corrosive	HP3—Flammable; HP4—Irritant; HP5—Specific Target Organ Toxicity (STOT); HP6—Acute Toxicity; HP7—Carcinogenic; HP8—Corrosive; HP10—Toxic for reproduction; HP11—Mutagenic
Zn/Mn	HP3—Flammable	HP3—Flammable; HP4—Irritant; HP5—Specific Target Organ Toxicity (STOT); HP6—Acute Toxicity; HP10—Toxic for reproduction; HP11—Mutagenic
Generic BM	HP3—Flammable	HP3—Flammable; HP4—Irritant; HP5—Specific Target Organ Toxicity (STOT); HP6—Acute Toxicity; HP10—Toxic for reproduction; HP11—Mutagenic

Table 12. Overview of technologies for recycling BM.

Technology	Description	BM Type	Recovered Elements	TRL	Limitations	Sources
Electrochemical junction transfer (ETJ)	A cation recovery process using intercalation into a host matrix, with a focus on lithium recovery from spent Li-ion batteries leachate.	Spent LIBs leachate	Lithium (close to 100% faradic yield after thermal treatment and pH adjustment)	TRL4-5	Presence of organic compounds and leachate acidity can block transfer; requires thermal treatment and pH adjustment	[44]
Mechanochemical treatment and acid leaching	Utilizing reductive reagents in milling for crystal structure modification and subsequent leaching with acid.	From e-bike LIBs	Lithium (maximum extraction of 29.9% in water leaching), nickel extraction improved from 78% to 92% with CoS in acid leaching	TRL4-5	No significant improvements in crystal structure post-milling; pervasive fluoride presence	[45]
Mechanical Activation-Assisted Pyrometallurgy	Employs graphite in the black mass to produce Fe/Cu-based alloys, adding metal oxides (Fe ₂ O ₃ , CuO) to BM then using mechanical activation to enhance efficiency.	From LCO and NMC LIBs	Specific rates not de-tailed	TRL4-6	High energy consumption in traditional pyrometallurgy; specific challenges with reaction kinetics and temperature control; potential issues with final alloy composition and purity	[29]
Combined pyro- and hydrometallurgical process	Electric arc furnace smelting to generate lithium-enriched slag and mixed metal alloys.	From spent LIBs, treated pyrometallurgically	Lithium (82.4% yield in slag), cobalt (81.6%), nickel (93.3%), copper (90.7%)	TRL3-5	High-temperature processing complexity	[22]
Hydrometallurgical recovery from lithium slag	Direct leaching of lithium slag in H ₂ SO ₄ and dry digestion method to reduce silicon gel formation.	Pyrolyzed LIBs BM	Lithium (close to 100% efficiency after 30 min at 20 °C), Al and Si (variable efficiency)	TRL3-5	Silicon gel formation complicating filtration and recovery	[23]
Early-stage lithium recovery (ESLR) using thermal treatment and supercritical CO ₂ leaching	Lithium recovery shifted to the start of the chemo-metallurgical process, with CO ₂ supercritical state enabling selective lithium leaching.	From NCM-based electric vehicle cells, thermally treated	Lithium (up to 79% yield with supercritical CO ₂ treatment)	TRL3-5	Complexity in managing lithium yield influencing factors, such as pyrolysis temperature and autoclave carbonation setup	[25]
Acid leaching using molasses as reductant	Utilizes acetic acid and molasses for metal leaching from BM, reducing higher oxidation states of metals.	From spent cylindrical lithium-ion batteries	Co, Li, Ni, Mn (96% to 99% recovery efficiency)	TRL5-6	Balancing molasses concentration and timing; sediment formation management	[26]
Carbothermic reduction and hydrochlorination	Involves reducing metallic oxides to metals, converting Li and Mn into soluble chlorides, and Co and Ni into magnetic alloys. The Al remains in the form of Al ₂ O ₃ and does not react.	From spent lithium-ion batteries	Li (97.28%), Mn (98.13%), Ni and Co in magnetic fraction (93.03%, 91.37%), Al in non-magnetic fraction (95.28%)	TRL5-6	Managing material streams, specific separation requirements, precise process condition control	[27]
Direct selective leaching of lithium.	Selective lithium leaching from BM using formic acid and hydrogen peroxide, allowing for targeted extraction while leaving other metals.	Industrial BM from Li-ion batteries with LiFePO ₄ cathodes	Lithium > 97%; other metals (Fe, Cu, Al, Ni, Co, Mn) < 1% leaching	TRL5-6	Need for operational condition optimization	[28]

Table 12. Cont.

Technology	Description	BM Type	Recovered Elements	TRL	Limitations	Sources
NaOH Leaching	Targets separation of active cathode material by selectively leaching the aluminum cathode electrode with an alkaline solution to yield a relatively pure black mass.	Derived from cathode material of LIBs	Al: 90.54%; Co: 99.95%; Li: 99.80%; Mn: 99.98%; Ni: 99.98%	TRL3-5	Inability to completely remove the protective Al ₂ O ₃ layer; dilute quantities of valuable metals in leach filtrate; potential losses of Co, Li, Mn, and Ni during the process	[40]
Use of organic solvents (DMF, DMAc, DMSO) and alkaline solutions (NaOH, KOH).	Organic solvents used are DMF, DMAc, DMSO; alkaline solutions are NaOH and KOH.	Mixed chemistries from LIBs	Lithium efficiency improved from 47.2% to 78.7% with DMSO; Cobalt efficiency improved from 28.5% to 61.3% with DMSO; Lithium extraction up to 90.1% with NaOH; Cobalt extraction up to 74.4% with NaOH	TRL5-6	High temperatures (>150 °C) for organic solvents; concentrated alkaline solutions pose corrosion and handling risks	[46]
Leaching of industrial BM.	Leaching with a solid-liquid ratio of 1:10 or 1:20 g/mL.	From spent LIBs	Co from LCO ~32%; Li from LCO ~63%; Li, Mn, Ni 100%; Co 100% after 60 min; Al 68% after 60 min; Cu 98% after 5 min then decreases to 60%	TRL5-6	Operational conditions optimization needed	[47]
Solvent extraction for Zn and Mn recovery.	Solvent extraction using ionic liquids, organophosphorus-based acids, and deep eutectic solvents (DESs).	From spent Zn-MnO ₂ alkaline batteries	100% for Zn (II) with (Cyanex 272 + diethyl phosphite); 100% for Mn (II) using DES	TRL5-6	Further studies needed for process optimization and extraction efficiency improvement	[48]
Solid-liquid-liquid extraction	Extraction of heavy metals from Ni-Cd battery BM using deep eutectic solvents (DESs) and other extractants in toluene or naphtha.	From spent Ni-Cd batteries	Ni (II): 30 wt.%; Cd (II): 100 wt.%	TRL 4-6	Lower extraction efficiency for Ni compared to Cd; complexity in the extraction process	[33]
Electro-assisted leaching	Involves electro-assisted leaching for metal recovery from BM, followed by electrochemical deposition.	From dismantled Ni-Cd batteries	Cd: High recovery rate; Ni and Co: Slower dissolution, specific rates not detailed	TRL 4-6	Proton generation control at the anode, slower dissolution of Ni and Co compared to Cd, high energy consumption, complex process design and operation	[49]
Extraction with Ionic liquids (ILs), deep eutectic solvents (DESs), organophosphorus-based acids	Low-temperature method for extracting cobalt, nickel, lithium, and other metals from spent LIB BM without pre-leaching.	From spent lithium-ion batteries	Co (II): 90–100 wt.% using DESs; Li(I): Up to 100 wt.; Ni (II): Up to 52 wt.%	TRL 4-6	Complex extraction process, variability in recovery rates based on methods and conditions	[30]
selective sulfation roasting	Extracting cobalt and lithium from LCO-rich BM using selective sulfation roasting, followed by water leaching.	From industrial LCO-rich spent batteries	Co: Up to 61.21%; Li: Up to 99.51%; Ni: Up to 33.00%; Mn: Up to 68.36%; Cu: Up to 24.53%	TRL 4-6	Dependency on roasting conditions and carbon presence, complexity in process design and operation	[31]
Scalable direct recycling of cathode BM from spent LIBs	Integrates pretreatment and relithiation of cathode BM from EoL LIBs.	From EV batteries, specifically NCM cathode materials	100% electrochemical performance recovery, 91% yield rate, regenerated cathode material exhibits 82% ICE with 176 mAh/g discharge capacity, 94% capacity retention after 200 cycles	TRL 6-7.	Precision control required for hydrothermal and annealing conditions, efficient removal of impurities	[50]

The construction of a European infrastructure capable of efficiently managing the current and future amount of BM becomes a compelling economic proposition. In light of Europe's strategic imperative to internalize BM recycling processes, a combined technical and economic analysis of the prevailing technologies is crucial. This analysis must weigh the operational maturity and recovery efficacy of each process against its economic viability and alignment with sustainable development objectives. Emerging recycling technologies, such as direct leaching in H_2SO_4 with a dry digestion method [23], acid leaching using molasses [26], and selective leaching using formic acid and hydrogen peroxide [28], are distinguished by their high-efficiency rates, achieving up to 100% lithium recovery. These methods signify considerable strides in material recuperation from expended LIBs. However, the economic feasibility of such high-efficiency technologies must be gauged considering the significant initial capital investment, operational expenditure, and maintenance requisites. The fiscal implications of achieving such high recovery rates must justify the financial and environmental costs incurred. Furthermore, innovative processes like electrochemical junction transfer (ETJ) [44] and early-stage lithium recovery (ESLR) using supercritical CO_2 leaching [25], despite being at the vanguard of physicochemical innovation, require enhancements in energy efficiency and CO_2 emission mitigation. These enhancements are essential to ensure their economic and environmental sustainability. The operational complexities of technologies like carbothermic reduction and hydrochlorination [27] and electro-assisted leaching [49], while promising in recovery rates, necessitate rigorous management of materials and fine-tuning of process conditions. The economic implications of these processes must account for their maintenance intensity and the requirement for continuous adaptation to evolving battery chemistries. The mechanochemical treatment and acid leaching approach [45], although less energy-demanding, highlights the necessity for process flexibility in the face of persistent impurities. This underscores the fact that the economic viability of recycling technologies is contingent upon the specific composition of the battery materials being processed. Methods such as solvent extraction for Zn and Mn recovery [48] and the exploitation of organic solvents [46], positioned at TRL 5-6, represent a critical juncture in the transition from experimental to scalable operations. These processes must maintain a delicate balance between the elevated operational temperatures required and the imperative for optimization to be economically viable. Lastly, the scalable direct recycling of cathode BM [50] promotes a recycling process that adheres to circularity principles. This not only ensures high recovery rates but also maintains the functional integrity of the material, offering a sustainable trajectory for recycling that could have a profound economic impact.

5. Conclusions

The present study has delved into the multifaceted and pressing issue of BM recycling from batteries, an area growing in urgency alongside the expected increase in end-of-life batteries. The research has unveiled the inherently complex and hazardous character of BM, as deduced from the interplay of various regulations within the European framework that govern the categorization and labeling of substances and waste. A pivotal finding of this inquiry is the indispensable need for definitive legislative action to classify the hazardous nature of BM, and in this regard, the study contributes significantly by establishing that BM should be classified, at a minimum, under the HP3 due to its flammability, while also acknowledging its relevance to a broader spectrum of HPs.

Addressing the identified hazards, the study concurrently casts light on the emergence of innovative technologies and methodologies that promise to mitigate these risks while bolstering the efficiency of BM recycling—answering the second research question. These advancements lend credence to the notion that while BM recycling presents formidable challenges, they are surmountable with the development and application of such technologies, which are vital to revolutionizing BM handling and enhancing the safety and efficacy of recycling practices.

While this research has imparted significant insights, it also delineates avenues for further investigation. The limited sample size, while illustrative, invites more comprehensive future studies employing the robust methodology established here to deepen the understanding of BM recycling across a more extensive range of BM types and compositions.

The study also acknowledges the complexity introduced by the variability of BM composition across different battery types. This diversity, though challenging, offers fertile ground for further research into an array of recycling methods, potentially yielding more nuanced and efficient recycling strategies that accommodate the vast spectrum of BM characteristics.

As Europe faces an influx of battery waste and an accompanying rise in BM volume, this study identifies an urgent need to appraise the continent's current recycling capacities and to pinpoint existing gaps. Addressing these deficiencies calls for an integrated approach in investing and evaluating the innovative technologies explored in preceding chapters. Notably, those technologies at a higher TRL—such as acid leaching using molasses, scalable direct recycling, and direct selective leaching of lithium—showcase operational maturity and a high degree of material recovery efficiency, suggesting their potential for scalable and economically viable applications.

Future research endeavors should endeavor to perform a detailed economic feasibility analysis of these technologies, considering their potential for industrial-scale application, the time required for technological maturation, cost implications, and the possible revenues from the recovered materials. Such an approach is essential to assure that the recycling processes are economically appealing and environmentally prudent, attracting investment and fostering entrepreneurial interest.

The conducted study establishes a pivotal foundation for future advancements in BM recycling. Bridging scientific inquiry with legislative frameworks, it marks a significant stride towards responsible and effective management in the battery recycling domain. This investigation not only tackles the immediate complexities but also establishes a solid base for future scientific and regulatory developments, which are vital for navigating the environmental challenges ahead.

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Abbreviations

Al	Aluminum
As	Arsenic
BM	Black Mass
Cd	Cadmium
Ca	Calcium
C	Carbon
CSBM	Category-Specific Black Mass
CLP	Classification, Labeling, and Packaging
Co	Cobalt
Cu	Copper
DES	Deep Eutectic Solvents
ESLR	Early-stage Lithium Recovery
ECHA	European Chemicals Agency

EC	European Commission
ETJ	Electrolytic Junction Transfer
EU	European Union
F	Fluorine
GHS	Globally Harmonized System
HSC	Harmonized System Codes
HP	Hazardous Properties
IL	Ionic Liquids
Fe	Iron
Pb	Lead
LIB	Lithium-Ion Batteries
LCO	Lithium Cobalt Oxide
Mg	Magnesium
Mn	Manganese
Ni	Nickel
NMC	Nickel Manganese Cobalt
P	Phosphorus
K	Potassium
PRISMA	Preferred Reporting Items for Systematic Reviews and Meta-Analyses
Si	Silicon
Ag	Silver
Na	Sodium
STOT	Specific Target Organ Toxicity
SLR	Systematic Literature Review
TRL	Technology Readiness Level
Sn	Tin
Ti	Titanium
Zn	Zinc

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