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Material Flow Analysis of Lithium-Ion Battery Recycling in Europe: Environmental and Economic Implications †

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Abstract: This study aimed at a quantitative analysis of the material flows associated with End of Life (EoL) lithium-ion batteries' (LIBs) materials in Europe. The European electric vehicles fleet in 2020 was taken as a case study, assuming a 10-year lifetime for the batteries and that the related EoL LIBs would be processed by existing recycling plants via pyrometallurgy, hydrometallurgy, or their combination in sequence. The economic implications (recycling operative costs compared to the revenues from the sales of the recycled metals) and the environmental performances (CO_2 eq. emitted, energy demand and circularity performances) were assessed. Based on the gathered results, the existing European recycling capacity will overlook over 78% of the forecasted EoL LIBs. The treatment efficiencies of the full-scale recycling processes allow for the recovery of over 90% of copper, cobalt, nickel, and manganese, 87% of aluminum, and only 42% of lithium and 35% of iron entering the recycling facilities. In overall, LIBs recycling in 2030 will involve the emission of 3.7 Mt of CO_2 eq. and an energy demand of 33.6 GWh. Hydrometallurgy presents the best economic and environmental trade-off compared to other recycling strategies. In conclusion, this study demonstrated that current European LIBs' recycling infrastructure will be inadequate in the near future and the direction (i.e., hydrometallurgy) that its strengthening should pursue.

Keywords: circular economy; economic analysis; environmental analysis; lithium-ion battery; material flow analysis; recycling

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1. Introduction

Lithium-ion batteries (LIBs) are a developing technology for energy storage that finds numerous applications from electric vehicles (EVs) to portable electronic devices (laptops, smartphones, tablets, etc.). LIBs are a key flexible option for the success of the energy transition [1], and they stand as a fundamental technology in the decarbonization of transportation and energy systems. Nonetheless, the sustainability of the whole LIBs' life cycle is strongly dependent on the management of their end-of-life (EoL) stage because they generate hazardous wastes associated with environmental and health safety issues and, on the other hand, represent a resourceful potential mine for secondary raw materials. LIBs' composition includes several economically valuable elements and critical raw materials (CRMs), whose mining activities are often associated with significant costs and environmental impacts [2]. In details, several LIB components, such as lithium, cobalt, phosphorous, and graphite, are listed as CRMs by the European Commission (EC) [2]. LIB recycling is essential to mitigate raw materials' shortage and the economic and environmental costs related to their primary extraction. Indeed, current legislations at the European level are targeting LIBs end of life management in order to curb environmental criticalities, requiring separate collection [3] and encouraging strategies to extend LIBs life cycle and close materials loop [4]. Although LIBs recycling is strongly encouraged by several policies [5,6], policies concerning extended producer responsibility represent a particularly important driver for EoL LIBs material, flowing towards scenarios more reliant on recycling strategies [7]. Presently, LIBs' recycling at full-scale relies on pyrometallurgical and hydrometallurgical processes allow for recovering the most valuable battery components, e.g., the metals present in the electrodes [8], which could potentially replace primary raw materials in the manufacture of electric vehicles [9]. Pyrometallurgical treatments consist of high-temperature reduction processes, characterized by intense energy demand, leading to metallic alloys containing cobalt, iron, nickel, and manganese, while lithium is lost in the residual slags [10]. The phase and morphological transitions that take place at elevated temperature in the controlled atmosphere of pyrometallurgical reactors allow us to decompose the organic components of the spent batteries and mainly purify the metals present in the cathodes as alloy, leaving Li in the reaction slag [11]. Furthermore, the degradation of the organic components present in EoL LIBs, such as binders, electrolyte, and acetylene black, generates toxic gasses emissions, for which the treatment system strongly affects the operative costs of LIBs recycling plants [12]. Hydrometallurgical processes are based on acid leaching followed by the precipitation of valuable metals such as salts or hydroxides, which serve as precursors for the secondary raw materials included in new batteries [8]. Hydrometallurgy is mainly applied at full scale as leaching with sulfuric acid followed by recovery of leached metals through chemical precipitation or electrodeposition [11].

The main drawback from acid leaching processes are the release of harmful compounds, such as Cl_2, SO_3, and NO_x and the generation of acidic waste water [13]. Therefore, recent studies are investigating the possibility of replacing conventional with organic ones, such as citric acid, malic acid, and succinic acid [11].

In this framework, since LIBs' recycling allows to recover secondary raw materials that can be re-introduced in the battery life cycle, curbing environmental and economic impacts. A virtuous example of materials circularity in the energy sector was posed by Lead batteries recycling. The European collection rate and recycling capacity reached 99% and 95% in the last decade, ensuring closed-loop circularity [14], turning recycled Lead scraps into the major source of lead batteries supply chain [15].

Similarly, the material flow analysis (MFA) of LIBs' recycling has been gaining increasing attention from the scientific community in recent years (Figure 1). The MFA studies describing LIBs' recycling published in the last five years (Figure 1A) mostly considered material flows related to EVs' market, while research on batteries for stationary energy storage [16] or portable electronics batteries [17–19] is still limited. This may be due to the fact that electrification of transport system is currently the main LIBs' application [20]. A relatively low number of studies (Figure 1B) applied MFA to a global perspective [21,22], whereas most studies considered specific geographic regions, e.g., China [23–28], South Korea [29], and Thailand [19] in Asia; The United State of America [28,30] and Brazil [31] in the American continent; and Austria [17], The Netherlands [32], Switzerland [16], and the United Kingdom [33] in Europe. Besides, some studies targeted only specific material flows, such as lithium [21,30], cobalt [34], or graphite [26], or specific aspects—e.g., criticality due to raw materials' shortage and supply risks [34,35] or environmental aspects [16,36].

To our knowledge, a specific study analyzing the material flows related to full-scale LIBs' recycling across Europe and the associated environmental and economic implications is still missing. The European Environment Agency [37] reported that between 2018 and 2020 the new registrations of EVs in EU-27 increased from 2% to 11% of circulating vehicles, and the forecast for 2021 involves a further increase to 18%. Considering an expected lifetime equal to 10 years for a LIB, the key research question posed by this study is "Will Europe be ready to recycle the EoL LIBs reaching the recycling plants in 2030?" This study has several elements of novelty and objectives: (i) a material flow analysis of the EoL LIBs forecasted in 2030, potentially deriving from the European EVs' fleet in 2020, and assuming a 10 year lifetime for the batteries; an assessment of (ii) the environmental (accounting CO_2 eq. emissions associated to energy needs, and circularity performance indicators), and of (iii) the economic implications of the material flows related to European EoL LIBs' recycling capacity in 2030, considering the different recycling strategies applied at full-scale.

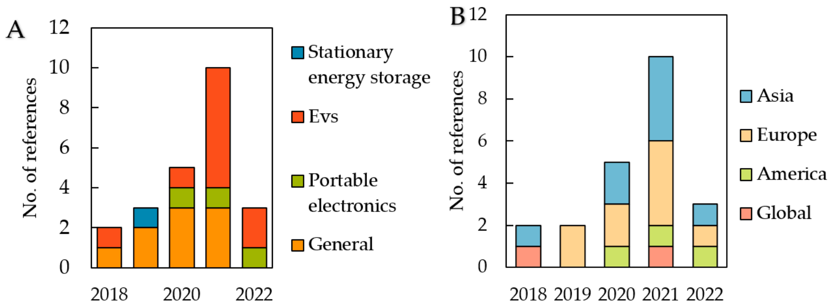

Figure 1. Overview of recent (2018–September 2022) literature studies about material flow analysis for LIBs' recycling clustered by (**A**) battery applications and (**B**) geographical boundaries.

2. Materials and Methods

2.1. Material Flow Analysis

This study accounted 720,913 EVs registered in Europe (EU-27, Iceland, Norway, and the United Kingdom) in 2020 [37], assuming an expected LIBs' lifetime equal to 10 years [38], and an average weight per battery equal to 318 kg [39]. Table 1 shows the market shares [40] and LIBs' compositions (%-wt.) [41] considered in this study. From these data, the amount (t) of materials associated with the European EVs' fleet in 2020 and the related amount (t) of waste materials reaching the recycling facilities in 2030 have been calculated [31]. The European LIBs' recycling capacity was assessed based on the current full-scale facilities (Table 2), and on the materials recovery efficiencies of the applied recycling processes (pyrometallurgy and hydrometallurgy) (Table 3) reported by the literature [32]. According to the processes applied in the inventoried European recycling facilities (Table 2), three recycling strategies have been considered in this study: pyrometallurgy, hydrometallurgy, and pyrometallurgy combined with hydrometallurgy. The recycling efficiency associated with pyrometallurgy combined with hydrometallurgy was assumed to be equal to the one of hydrometallurgy (Table 3); their combination allows us to recover lithium and iron, which are usually lost in the slag of pyrometallurgy as a single process [10]. Materials flow analysis (MFA) was performed using STAN2WEB open access software (version 2.6.801, http://www.stan2web.net, accessed on 21 March 2023) developed by Technische Universität of Wien according to the Austrian standard ÖNorm S 2096 (Material flow analysis application in waste management).

2.2. Environmental Analysis

The overall circularity of the material flows' management and the efficiency of the European recycling infrastructure have been estimated by defining three key performance indicators, as follows. In detail, the recovery rates related to the amount of generated waste (R_{TOT}) and of treated wastes (R_{TREAT}), and the efficiency of materials' use in the recycling processes ($\eta_{MATERIALS}$) have been calculated according to Equations (1)–(3), where R is the amount of recovered materials, G is the amount of wastes produced, T is the amount of treated waste, and I is the input of reagents and other materials employed in the recycling processes.

$$R_{TOT}(\%) = \frac{R(t)}{G(t)} \cdot 100 \tag{1}$$

$$R_{TREAT}(\%) = \frac{R(t)}{T(t)} \cdot 100 \tag{2}$$

$$\eta_{MATERIALS}(\%) = \frac{R(t)}{T(t) + I(t)} \cdot 100 \tag{3}$$

Table 1. Market shares and compositions of the LIBs (identified by the different cathodes) considered in this study (CC: current collector; DMC: dimethyl carbonate; EC: ethylene carbonate; LFP: lithium iron phosphate; NCA: nickel cobalt aluminum; NMC: nickel manganese cobalt; PVDF: polyvinylidene fluoride; PE: polyethylene; PET: polyethylene terephthalate) [40,41].

Battery		LFP	NCA	NMC 111	NMC 622	NMC 811
Market Share		2%	19%	24%	45%	10%
Composition (%-wt)	PET	0.4%	0.3%	0.3%	0.3%	0.4%
	PE	0.2%	0.3%	0.3%	0.3%	0.3%
	Electrolyte DMC	9.3%	6.3%	6.2%	6.3%	7.2%
	Electrolyte EC	9.4%	6.3%	6.2%	6.3%	7.2%
	Electrolyte LiPF6	3.3%	2.3%	2.2%	2.2%	2.6%
	Al (CC)	7.5%	8.4%	8.2%	8.4%	8.0%
	Cu (CC)	14.5%	16.9%	16.4%	16.8%	15.7%
	Binder PVDF	2.7%	2.9%	2.9%	2.9%	3.6%
	Carbon black	2.2%	2.1%	2.3%	2.1%	1.7%
	Graphite	16.6%	22.0%	19.0%	20.7%	20.6%
	Li	1.4%	2.2%	2.7%	2.5%	2.4%
	Co	-	2.8%	6.9%	3.8%	1.9%
	Ni	-	14.9%	6.9%	11.5%	14.9%
	Mn	-	-	6.4%	3.6%	1.7%
	Al	-	0.4%	-	-	-
	Fe	11.4%	-	-	-	-
	P	6.3%	-	-	-	-
	O	13.1%	10.1%	11.2%	10.4%	10.2%

Table 2. Inventory of European LIBs' recycling capacity (n.s.: not specified).

Country	Company	Pyrometallurgy (t/y)	Hydrometallurgy (t/y)	Reference
Belgium	Umicore	7000	7000	[42–44]
Finland	Akkuser	4000	-	[43]
France	SNAM	300	-	[42–44]
	Recupyl	-	-	[8,43,44]
	Euro-dieuze	-	200	[43]
	Eramet	20,000	-	[43]
Germany	Accurec GmBH	4000	4000	[8,42]
	Duesendeld	3000	-	[43]
	Redux	-	10,000	[42]
	Lithorec	-	n.s.	[8]
Spain	Pilagest	-	-	[45]
Sweden	uRecycle	-	-	[43]
Switzerland	Batrec Industrie AG	-	200	[8,44]
United Kingdom	AEA Technology	-	n.s.	[8,42,44]

Table 3. Recovery efficiencies achieved by different recycling technologies [8–12,38,42,44].

Target Element	Pyrometallurgy	Hydrometallurgy
lithium	-	95% ± 7.0%
cobalt	86% ± 15.0%	95% ± 5.8%
nickel	98% ± 1.0%	97% ± 3.2%
manganese	88% ± 4.0%	91% ± 19.7%
aluminum	99%	71% ± 31.1%
iron	-	80% ± 20.9%
copper	96%	95% ± 11.2%

Moreover, the environmental consequences of current recycling strategies applied at full scale across Europe have been estimated in terms of Green House Gasses (GHGs) emissions, related to different recycling strategies. Operative parameters of different recycling strategies have been considered as energy consumption, required reagents, and wastes generated by the processes. Their values were obtained from Ecoinvent 3.8 database [46]. The greenhouse gas specific emissions associated with the energy demand of the applied LIBs' recycling strategies, equal to 110 kg CO_2/kWh, have been accounted based on the average European energy mix forecasted for 2030, when the EU-27 energy mix will account for at least 30% of renewable energy sources [47].

2.3. Economic Analysis

The economic analysis assessed the trade-off between the recycling processes' operative costs and the market values of the recovered material streams (i.e., the revenues). Markets values considered as costs and revenues have been defined according to Ecoinvent 3.8 database. The considered revenues were limited to the sales of the recoverable metals entailed in cathodes and current collectors (0.81 €/kg for aluminum, 3.6 €/kg for copper, 6.75 €/kg for lithium, 10.4 €/kg for cobalt, 1.24 €/kg for manganese, 1.5 €/kg for iron, and 1.0 €/kg for phosphorous). These values were then estimated based on the number of treated materials and recovery efficiency of the recycling processes, as defined in the material flow analysis. The considered operative costs (average for EU-27) were related to energy (0.086 €/kWh) [48], chemicals employed in the recycling treatments (0.19 €/kg for sodium hydroxide, 0.06 €/kg for sulfuric acid, and 0.45 €/kg for lime), and to the landfill fees for the disposed waste (22.0 €/t) [49]. Other costs (mechanical pre-treatments energy demand, labor, maintenance, etc.) were excluded, as they could be assumed as equivalent compared to the recycling treatments [11]. The overall potential revenues and treatment costs have been normalized by the mass of treated materials to compare the different processes.

3. Results

3.1. Material Flow Analysis

This study considered the European EVs' fleet in 2020, based on the different market shares of commercially available battery chemistries and their compositions, and 229 kt of EoL LIBs are expected by 2030. In detail, 103 kt of NMC622, 55 kt of NMC111, 44 kt of NCA, 23 kt of NMC811, and 4 kt of LFP EoL batteries are foreseen. These also imply 5.7 kt of plastics (polyethylene and polyethylene terephthalate), 34.7 kt of electrolytes (dimethyl carbonate, ethylene carbonate and LiPF6), 6.8 kt of organic binder, 46.9 kt of graphite, 38 kt of copper, 19 kt of aluminum, 5.6 kt of lithium, 9.3 kt of cobalt, 25.5 kt of nickel, 7.6 kt of manganese, 0.5 kt of iron, and 0.3 kt of phosphorous. This study concerned the metals contained in cathodes, holding the majority of EoL LIBs' economic value [8].

According to the inventory of the treatment plants (Table 2) performed to complete the MFA, European LIBs' recycling capacity amounts to 48.8 kt, where 56% of input materials (27.3 kt) can be treated by pyrometallurgy, 21% (10.3 kt) can be treated by hydrometallurgy, and 23% (11.2 kt) can be treated by pyrometallurgy followed by hydrometallurgy. The material flows associated with EoL LIBs forecasted for 2030, based on the European EVs' sales in 2020 and compared to European recycling capacity, are displayed in Figure 2.

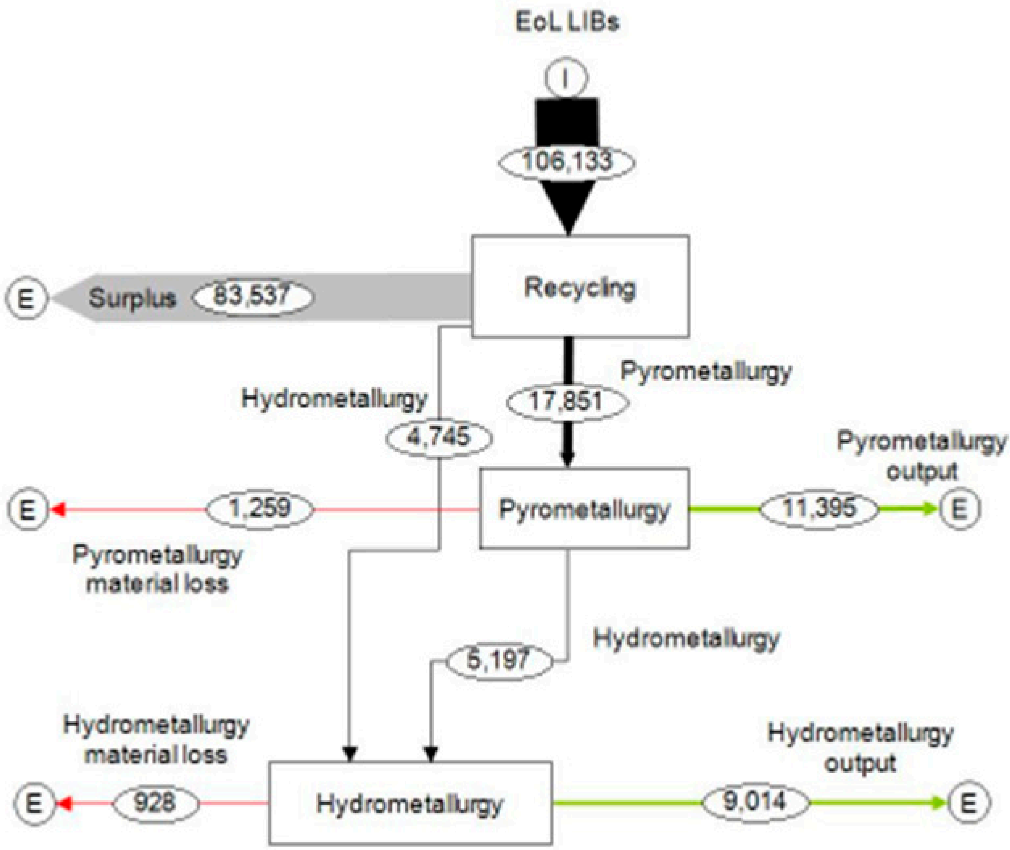

Figure 2. Material flow analysis of European electric vehicles fleet that will reach end of life in 2030 and performance of European LIBs recycling (data are expressed in k tons) (I: input flow, E: exit flow).

Pyrometallurgy is the most applied technology in full scale recycling plants (Table 2). Thereby, it is evident that the potentially recoverable secondary raw materials associated with pyrometallurgy are higher (+26%) compared to hydrometallurgy, and the same happens with material losses. Moreover, it should be noticed the inadequacy of the European LIBs' recycling infrastructure compared to the waste streams expected in 2030, i.e., almost 230 kt of materials deriving from EoL batteries, is expected to reach EoL stage, and that will correspond to an amount 3.7 times higher than the existing recycling capacity. According to the results of the MFA (Figures 2 and 3), the total forecasted input of recoverable metals (aluminum, copper, cobalt, manganese, iron, lithium, and phosphorous) entailed in cathodes and current collectors of the EoL LIBs entering the European recycling plants in 2030 will be equal to 106,133 t. These materials will be distributed across different facilities, up to the saturation of each plant recycling capacity (Table 2). According to the recovery efficiencies of the recycling processes applied at full scale (Table 3), the amount of recovered materials was estimated as: 3.5 kt of aluminum (87% of the forecasted recycling input), 7.8 kt of copper (96% of the input), 0.5 kt of lithium (42% of the input), 1.8 kt of cobalt (90% of the input), 5.3 kt of Nickel (98% of the input), 1.5 kt of manganese (90% of the input), and 39 t of iron (35% of the input). Most of the recovered metals will derive from pyrometallurgical processes—i.e., 64% of recovered aluminum, 56% of copper and nickel, 54% of cobalt, and 55% of manganese would be obtained through pyrometallurgy. On the other hand, the lowest recovery rates were observed for lithium (42% of the potentially recoverable amount) and iron (35%), which are lost in the slags during pyrometallurgy.

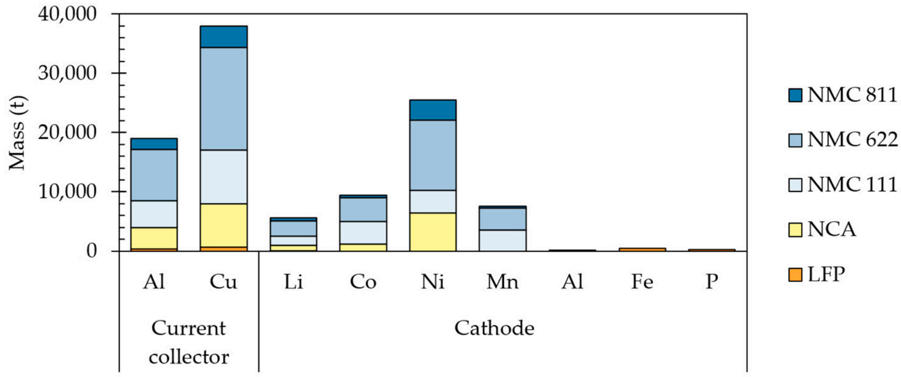

Figure 3. Amounts of potentially recoverable metals (t) entailed in LIBs' cathodes and current collectors associated to European EVs' fleet in 2020.

Current LIBs' recycling infrastructure has been proven effective in metal (lithium, cobalt, manganese, nickel, aluminum, iron, and copper) recycling, as it could potentially recover over 85% of cobalt, nickel, manganese, aluminum, and copper, and about 40% of lithium from the input materials. However, these rates are drastically diminished not only when the inputs to the treatment facilities are accounted for, but instead the amounts of materials are discarded as waste. Since the recycling infrastructure could be able to treat around 20% of EoL LIBs amount, the effective recovery rates will correspond to: 21% of nickel, 20% of copper, 19% of cobalt and manganese, 18% of aluminum, and less than 10% of lithium. Therefore, the bottleneck of LIBs' recycling is not the low efficiency of material recovery, since from the review of literature they appear to be above 80% for the above-mentioned metals (Table 3). Nonetheless, the lack of hydrometallurgical plants able to recover lithium and iron poses a considerable limitation to ensure full material circularity in the battery sector.

3.2. Environmental Analysis

The obtained values of the circularity indicators and recycling efficiency parameters (Table 4) showed that the recovery rates present a relevant gap when the total amount of generated waste is considered (as in R_{TOT}), rather than the amount of materials entering the treatment plants (as in R_{TREAT}), due to the high efficiency of the processes implemented at full scale and, conversely, the limited capacity of the overall recycling infrastructure. The recovery efficiencies (both R_{TOT} and R_{TREAT}) of lithium and iron will be particularly low (7–9% and 35–42%, respectively) due to the limited number of facilities implementing hydrometallurgical processes in Europe. Particularly, the observed gap between the performances of the recycling infrastructure associated to the input materials (R_{TREAT}) and to the overall EoL materials (R_{TOT}) should clearly indicate how the treatment infrastructure is not adequate to treat the EoL LIBs. Moreover, the efficiency in the use of materials ($\eta_{MATERIALS}$) in the recycling treatments will be particularly low for lithium (8%) and iron (1%), since they are mainly recovered through hydrometallurgy that, compared to pyrometallurgy, require a larger number of reagents for the leaching and recovery phases.

Table 4. Circularity indicators and recycling efficiency parameters (R_{TOT}: amount of generated waste; R_{TREAT}: number of treated wastes; $\eta_{MATERIALS}$: efficiency of materials use).

	Al	Cu	Li	Co	Mn	Ni	Fe	P
R_{TOT}	19%	20%	9%	19%	21%	19%	7%	19%
R_{TREAT}	87%	96%	42%	90%	98%	90%	35%	87%
$\eta_{MATERIALS}$	38%	58%	8%	25%	49%	21%	1%	38%

Based on the results of the MFA previously detailed, the overall GHG emissions associated with LIBs recycling in Europe in 2030 (3.7 Mt of CO_2 eq.) have been estimated according to the energy demand of different recycling strategies (pre-treatments, pyrometallurgy, and hydrometallurgy) applied at full-scale and to the average carbon footprint of energy production. The main contributor to the total energy demand of LIBs' recycling will be pyrometallurgy (21.84 GWh), due to its wide application across European plants. However, although almost the same number of materials will be treated by hydrometallurgy and combined pyrometallurgy and hydrometallurgy, the associated GHG emissions would be higher (10.34 GWh) for the combined processes compared to hydrometallurgy (1.46 GWh). These energy demands will entail about 3.7 Mt of CO_2 eq., accounting for 2.4 Mt of CO_2 eq. due to pyrometallurgy, 0.2 Mt of CO_2 eq. due to hydrometallurgy, and 1.1 Mt of CO_2 eq. due to combined pyrometallurgy and hydrometallurgy. Comparing these carbon efficiency values, the less emission-intensive option is hydrometallurgy, which is responsible for 16 kg CO_2 eq./t of treated material, whereas pyrometallurgy and pyrometallurgy, followed by hydrometallurgy, account for 88 kg CO_2 eq./t of treated material and 103 kg CO_2 eq./t of treated material, respectively. Moreover, other detrimental environmental consequences could stem from an unproper management of the untreated EoL LIBs exceeding the European recycling capacity, as LIBs' components present several pollution hazards [50], mainly due to the potential release of inflammable or corrosive substances or potentially toxic metals [51–53].

3.3. Economic Analysis

The economic analysis of the material flows involved by LIBs' recycling was performed considering the revenues derived from the sales of the recovered metals according to their market values (Section 2.3) and considering the recovery efficiencies of current full-scale applied technologies (Table 3), and the costs entailed by the recycling treatments (Section 2.3). Concerning the incomes (Figure 4), 22.6 kt of metals recovered through LIBs' recycling will correspond in total to 53,600,000 €, mostly due to cobalt (18,800,000 €), copper (27,700,000 €), lithium (3,400,000 €), and aluminum (2,900,000 €). The role of CRMs as cobalt and lithium in the economic analysis should be noticed. According to the output flows of the MFA, cobalt represents only about 9% of the total output. However, because of cobalt's critical market price, the associated economic benefits entail 35% of the revenues. Nonetheless, lithium (accounting for about 5 wt.% of the streams of recovered materials) holds a share of the revenues, which is limited up to 6% because its market values is still limited, despite the shortage in Europe.

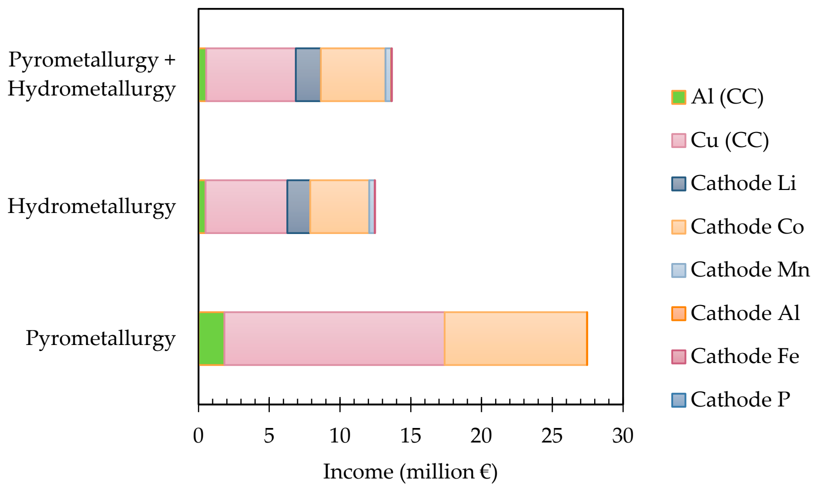

Figure 4. Potential incomes (million €) associated with the market value of the secondary raw materials involved by LIBs' recycling in 2030 (CC: current collector).

The costs entailed by LIBs' recycling processes (Figure 5) will be due to energy (particularly for pyrometallurgical processes) (Section 3.2), input materials, and landfill fees. The overall cost due to the energy demand of the full-scale treatment processes amounts to 2,890,000 €, mainly due to the large share of EoL LIBs (65%) treated in facilities applying pyrometallurgy (Table 2). While the number of materials treated by hydrometallurgy (21%) or by the combination of pyrometallurgy and hydrometallurgy (23%) does not differ significantly, the associated energy demand is considerably higher for the latter (about 31% of the 2,890,000 € spent for energy).

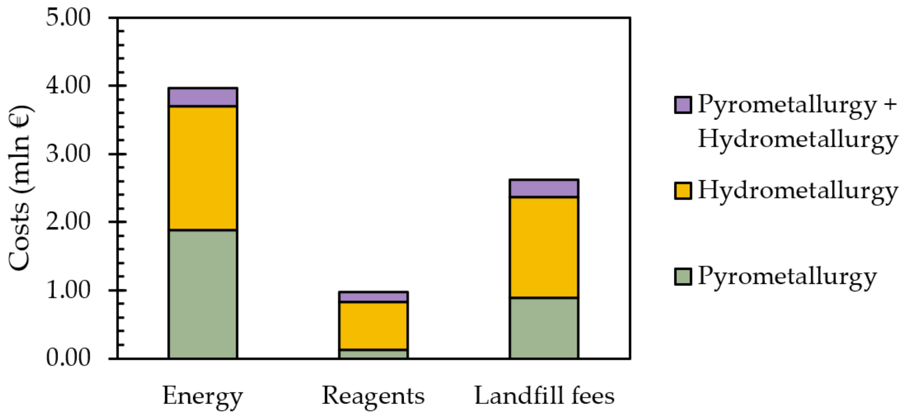

Figure 5. Costs (million €) associated to the material flows deriving from LIBs' recycling in 2030.

The input materials demand has been estimated as 13.3 kt of sodium hydroxide solution (71.5% by pyrometallurgical plants and 28.5% by pyrometallurgical plants with additional hydrometallurgical treatments), 5.3 kt of water (50.9% by pyrometallurgical plants, 13.2% by hydrometallurgical plants, and 35.9% by plants with pyrometallurgical and hydrometallurgical treatments combined), 4.9 kt of sulfuric acid, and 2.5 kt of lime for hydrometallurgical processes.

Consequently, the overall cost of materials required by the recycling processes will amount to 3,900,000 €, with 64% related to NaOH solution and 28% to lime, used in large quantities to precipitate metals from leaching solutions in hydrometallurgical plants. The costs related to landfill fees will be mainly due to the generation of residual non-Fe-Co metals and unrecyclable plastic mix. Specifically, each year, the current state of LIBs recycling infrastructure is expected to generate about 12.4 kt of low value metals scraps (without significant iron or cobalt content), 9 kt of waste plastic mix, and about 8.5 kt of other waste materials. About 700,000 € will account for landfill expenses, mainly due to disposal of non-Fe-Co alloys (entailing 41% of landfilling costs) and plastic mix (30% of landfilling costs).

Overall, considering the whole European LIBs' recycling infrastructure, the treatment of 48.8 kt of EoL LIBs will require 7,550,000 €, mostly (53%) due to pyrometallurgical plants, while 13% is ascribable to hydrometallurgy processes, and 35% is ascribable to combined pyrometallurgy and hydrometallurgy processes. The specific operating costs and potential revenues to treat a functional unit of 1 t of EoL LIBs have been estimated (Figure 6). The highest revenues are entailed by the plants where a combination of pyrometallurgy and hydrometallurgy is performed (1242 €/t), however this treatment generates the greatest operative costs (230 €/t). Moreover, considering the compromise between operative costs and potential revenues of the single treatments, pyrometallurgical processes, despite needing elevated operative costs (145 €/t), produce limited revenues (1006 €/t), mainly due to the materials loss in the slag phase, whereas hydrometallurgical treatments generate high revenues (1200 €/t) with the lowest operative costs (92 €/t). Eventually, considering the inadequacy of current recycling infrastructure proven in the MFA performed in this study, a wider system to collect and treat waste LIBs is essential. Moreover, according to the performances of existing recycling strategies (recovery efficiencies, energy demands with consequent emissions generation and cost-benefit trade-off), hydrometallurgy represents the best strategy.

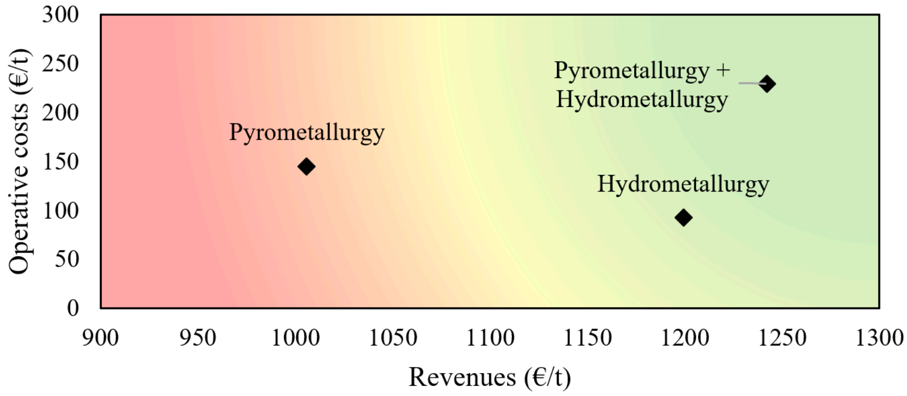

Figure 6. Comparison between operative costs and potential revenues of different LIBs' recycling strategies.

4. Conclusions

The existing literature analyzed the material flow analysis of valuable and critical materials across the LIBs' life cycle, highlighting the technical inadequacy of their recycling when it comes to non-cathodic metals. However, these studies seldom considered the economic and environmental implications of the recovery of secondary raw materials at full-scale in a large context like Europe. The present work aimed to assess the material flows potentially associated to the 229 kt of EoL LIBs reaching the European recycling plants in 2030, deriving from the EVs put into the market in 2020, and considering the treatment capacity of the existing recycling infrastructure. The results of the MFA confirmed the deficit of current European recycling infrastructure, due to the significant (around 80% of forecasted EoL LIBs) surplus that is not possible to treat in existing plants. Overall, the recovery rate of the treated materials is estimated to be over 85% for aluminum, copper, cobalt, manganese, and nickel, and between 35% and 42% for iron and lithium. Despite the high efficiency reached by up-to-date recycling technologies, the recovery of critical and secondary raw materials is still hindered by the wide-spread reliance on pyrometallurgy, applied in over 56% of the European recycling plants, which are not able to recover lithium. In 2030, the whole LIBs recycling infrastructure will imply the emission of 3,700,000 t of CO_2 eq. and an energy demand of 33.6 GWh (21.86 GWh required by pyrometallurgical plants, 1.46 GWh by hydrometallurgical facilities, and 10.3 GWh by the ones combining pyrometallurgy and hydrometallurgy). Considering the economic income as the net difference between potential revenues and operative costs, the best performance was presented by hydrometallurgical plants (1108 €/t), followed by plants with pyrometallurgical and hydrometallurgical processes in series (1012 €/t), and at last by pyrometallurgical plants (861 €/t). When considering specific costs and revenues, hydrometallurgy will entail the best trade-off. Indeed, although the potential revenues entailed by the combination of pyrometallurgical and hydrometallurgical treatments in series are the highest ones, considering the operative costs associated with LIBs recycling processes, hydrometallurgy is established as the best solution for battery recycling in terms of economic costs-benefits trade-off. Thereby, in view of the established inadequacy of current European recycling infrastructure, hydrometallurgy, as it is already well-developed at full-scale and is cost-effective, will pose a strategic recycling route for the design of future recycling facility. In conclusion, this study quantitatively demonstrated the inadequacy of current European recycling infrastructure to sustain the mass flows deriving from EVs LIBs expected in 2030. Moreover, hydrometallurgy was identified as the best recycling strategy from economic and environmental viewpoints.

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