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Material system analysis

A novel multilayer system approach to correlate EU flows and stocks of Li-ion batteries and their raw materials

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Abstract

Lithium-ion batteries (LIBs) will play a crucial role in achieving decarbonization and reducing greenhouse gases. If the EU wants to be competitive in the global market of LIBs, it has to ensure a sustainable and secure supply of the raw materials needed for the manufacturing of these batteries. Limited understanding of how the battery material cycles are linked with raw materials supply chains may hinder policy measures targeting the set-up of a domestic supply chain in the EU since no precise information on where to intervene will be available. The novelty of this work lies in a multilayer system approach developed to reveal interlinkages between the flows of five raw materials contained in LIBs (cobalt, lithium, manganese, natural graphite, and nickel) in the EU. This was achieved by aligning material system analysis datasets of raw materials contained in LIBs with datasets on stocks and flows of this type of batteries in the EU. The results demonstrate the EU's strong import dependency on LIBs and battery raw materials. The EU recycling of lithium and natural graphite is low/nonexistent hindering the sustainable supply of these materials. The results also show that the majority of battery materials are increasingly accumulated in use or hoarding stocks. The proposed approach is designed to help identify bottlenecks and possible solutions to increase the efficiency of the EU LIB system, which could go unnoticed if each material supply chain were examined individually. This study also highlights how the lessons learned can support EU resource-management policies.

KEYWORDS
industrial ecology, life cycle, Li-ion batteries, material flow analysis, raw materials, recycling

1 | INTRODUCTION

1.1 | Importance of Li-ion batteries and their raw materials

The transition to clean energy is expected to boost demand for batteries in the coming years. The European Commission has identified battery value chains as strategic industry elements in achieving European Union (EU) goals in terms of climate-neutrality and increasing competitiveness of EU industry (European Commission, 2019). The recent tightening of EU greenhouse gas (GHG) emission targets (President of the EC, 2020) is likely to accelerate the transition to achieve such goals.

LIBs technology is currently the global asset to achieve climate-neutrality through electrical mobility and stationary energy storage (European Commission, 2018a, 2018b). Since 2010, batteries used in electrical mobility and energy storage have shifted from nickel-hydride batteries and lead-acid batteries (LABs) to LIBs (Liu et al., 2021).

The EU is dependent on imports to satisfy its demand for LIBs. The world manufacturing capacity for LIBs cells is concentrated in Asia, namely China, Japan, and Korea. The same countries also dominate the upstream global manufacturing capacity for battery components: Cathodes (85%), anodes (97%), separators (84%), and electrolytes (64%) (Steen et al., 2017).

In the EU, the operating facilities manufacturing Li-ion cells are currently located in Hungary, France, Germany, Poland, and Sweden (Roskill, 2019; Steen et al., 2017). According to recent projections, Europe will increase its installed LIB manufacturing capacity from 48 GWh in 2020 to 670 GWh in 2030, before reaching 1100 GWh in 2040 (Fraser et al., 2021; Tsiropoulos et al., 2018).

The manufacturing of LIBs relies on several raw material resources for which the EU is also dependent on imports. If the EU wants to be competitive in the global market of LIB manufacturing, it must increase its capacity, which also depends on its ability to ensure the supply of the necessary raw materials (European Commission, 2018a). Limited understanding of how the battery material cycles are linked with raw material supply chains may hinder the EU's access to the battery market in the future. Therefore, monitoring material cycles for the most important battery raw materials alongside the battery supply chain is of great importance to the EU (Di Persio et al., 2020).

1.2 | Material flow analysis of Li-ion batteries

Material flow analysis (MFA) techniques using the mass conservation principle are powerful tools that provide crucial information on the flows and stocks of a target material within a given system. They can provide evidence to inform decision-making on the sustainable and competitive supply of battery raw materials (Müller et al., 2014).

The state-of-the-art on battery raw material flows does not facilitate the correct characterization of the LIB system in the EU since the majority of the publications take a global perspective (Hache et al., 2019; Harper et al., 2012; Schmidt et al., 2016; Simon & Weil, 2013; Sun et al., 2017; Talens Peiró et al., 2013; Ziemann et al., 2012) and there are only a few studies of LIB systems at a regional level (Asari & Sakai, 2013; Chang et al., 2009; Hao et al., 2017; Liu et al., 2021; Nigl et al., 2020; Song et al., 2019). Even fewer have the EU as a scope (Bobba et al., 2019; Lebedeva et al., 2017). The estimated dramatic increase in the demand for several battery-related raw materials for electric vehicle batteries and energy storage (European Commission, 2020a) will have a significant impact on the primary raw materials production routes especially of lithium (Hache et al., 2019; Olivetti et al., 2017; Pehlken et al., 2017). Secondary material sources, also known as anthropogenic stock or in-use stock, are considered to be key means of diversifying and securing resource supply, relieving reliance on imports, and ultimately strengthening the competitiveness and resilience of the European industry (Bobba et al., 2019; Schmidt et al., 2016; Ziemann et al., 2018). However, high dispersion in discarded products and waste, lack of incentives, relatively long product hoarding, and inefficient sorting and separation often result in significant losses at end-of-life and low recovery rates of raw materials through recycling. Developing strategies for end-of-life recovery and recycling is imperative and is built upon a thorough understanding of material cycles.

1.3 | Aim and novelty of the study

Most efforts to date have been limited to elemental cycles assessed individually, with flows and stock commonly expressed in metallic equivalent of a given resource (Chen & Graedel, 2012). Although this choice is better than simply using accounting procedures, it results in loss of information (Nakamura et al., 2017; Reuter et al., 2006) when, for instance, a metal under scrutiny is utilized in combination with other metals in selected applications, which can ultimately result in problem shifting. This aspect becomes relevant for the setting of material-recovery strategies at end-of-life as the quality of the recovered materials is also key to their further reuse and recycling (Nakamura et al., 2014; Ohno et al., 2014). Therefore, approaches that consider possible interconnections between different material cycles and in the LIB supply chain would provide a better
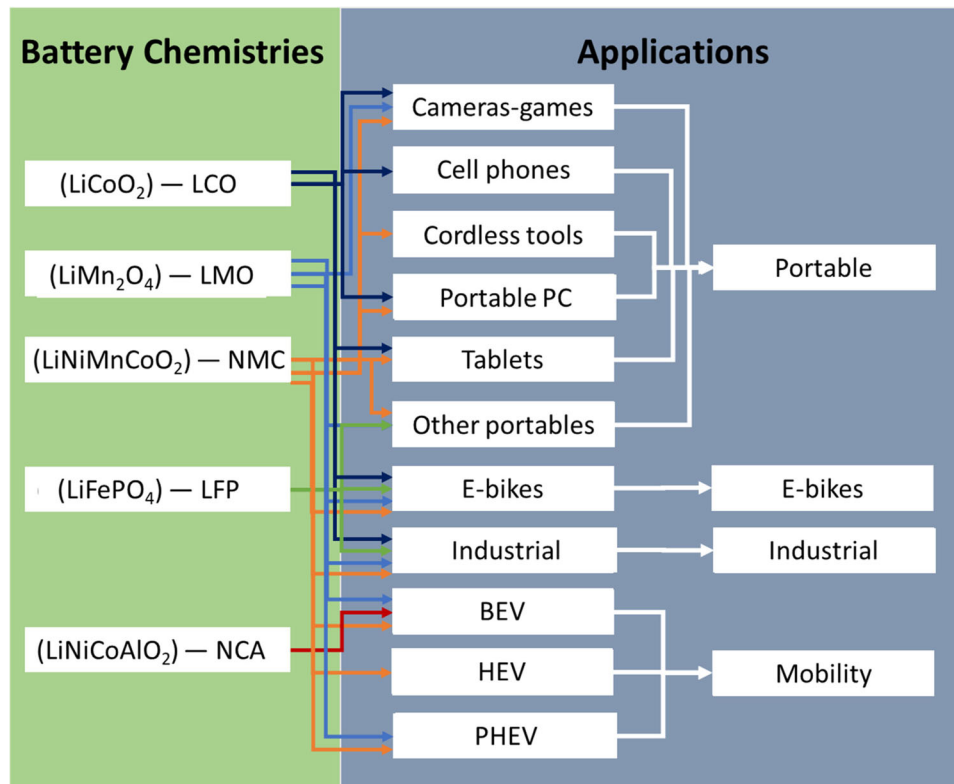
FIGURE 1 Battery chemistries and applications for the EU Li-ion rechargeable batteries system considered in the current study
Note: LMO-HEV and LFP-BEV also exist as does LFP for heavy transport (buses and e-trucks), but they were not relevant in the EU market in 2016. LFP cell-to-pack is the preferred choice for heavy duty right now. Other Li battery applications include primary batteries and batteries for heavy duty transport, but they were not included in this analysis

understanding of dynamics driving production and consumption of materials and goods (Ohno et al., 2016; Song et al., 2019). In particular, Song et al. (2019) demonstrate the advantages of analyzing simultaneously the life cycle of five critical raw materials for the Chinese LIB sector.

Building on individual anthropogenic cycles of five raw materials developed in related publications (Ciacci et al., 2021; Godoy León et al., 2021; Lundhaug et al., 2021), and following the recommendations of Hamilton in 2017 (Hamilton, 2017), this work develops and applies a multilayer system approach to: (i) linking layers of material cycles of five raw materials, namely cobalt, lithium, manganese, natural graphite, and nickel because of their essential presence in high-energy battery systems and (ii) analyzing the whole battery system across the EU LIB supply chain. The system approach developed in this study provides quantitative assessments of the links between the cycles of the materials and their applications that will identify bottlenecks and potential improvements for the EU to secure material supply and increase its competitiveness on the LIB market.

The comprehensive knowledge gathered by this study may support decision-making in the development of targeted strategies, policies, and investments for (i) raw material sustainable supply to the EU LIB sector, (ii) increasing resource efficiency (e.g., by identifying potential for improving the recycling performance of the selected metals), (iii) improving waste management, and (iv) supporting future research to address complementary and transversal challenges along the entire life cycle of the targeted resources. In particular, the possibility provided by the current analysis of examining the flows of the selected materials in all of their main applications identifies ways of securing secondary resources for the increasing LIB sector, once they are available at their end-of-life. The ongoing policies in the EU's Circular Economy Action Plan, the Strategic Action plan for Batteries (European Commission, 2018b), and the proposal for a new EU regulation on batteries and waste batteries (European Commission, 2020c) may benefit from the results of this paper. The multilayer system approach adopted here can be customized to target any EU sector as well as any material of interest.

2 | METHODOLOGY

2.1 | Materials, system boundaries, and temporal and technological coverage

Figure 1 shows the battery chemistries and their applications considered in this paper as part of the EU LIB system, with the focus being on Li rechargeable batteries. The applications studied include portable electronics (portable PCs, cell phones, cameras/games, tablets, cordless tools,

and other portables), electrical mobility (battery electric vehicles [BEV], plug-in hybrid electric vehicles [PHEV], hybrid electric vehicles [HEV]), e-bikes, and industrial batteries excluding mobility. This study follows the battery classification developed by Huisman et al. (2020) and RMIS (2019). Material flows were calculated for each battery application and for five raw materials contained in them: Cobalt (Co), lithium (Li), manganese (Mn), natural graphite (NG), and nickel (Ni). The material system analysis (MSA) studies were focused on the description of material flows of natural mineral raw materials. Therefore, the values reported for graphite are only representative of natural graphite, which represents around two-thirds of all the graphite used in LIBs. All of these raw materials, except nickel, were also mentioned in the Strategic Action Plan on Batteries (European Commission, 2018b) as priority raw materials under the pillar "secure access raw materials." The system boundary for the EU LIB and materials system, included manufacturing, use, collection, recycling, and trade. Extraction and processing stages of battery raw materials are discussed in the companion papers in great detail (Ciacci et al., 2021; Godoy León et al., 2021; Lundhaug et al., 2021). All the flows were calculated for the EU-28 and are representative of the year 2016. This was the year with the more complete dataset for all the materials analyzed. In addition, the evolution and trends of LIBs in the EU are also presented in Supporting Information S1 from 2012 to 2019 (Huisman et al., 2020).

2.2 | MSA and multilayer system

The study followed the MSA methodology developed for the European Commission with an EU scope in 2015 (BIO by Deloitte, 2015) and further revised in 2020 (Torres de Matos et al., 2020). The MSA is a quasi-stationary model MFA where the system border is all of the EU and where life cycle stages (including trade) are consistently defined for all raw materials. The main goal of an MSA is to serve policy needs for raw materials, such as the EU criticality assessment in order to draw up the list of critical raw materials (European Commission, 2020b) and to monitor the circular economy in order to calculate the end-of-life recycling input rate (EOL–RIR) (Eurostat, 2020). As an MFA, the MSA follows the general principles of mass conservation to map and quantify stocks and flows of raw materials along their life cycle in the EU, covering extraction, processing, manufacturing, use, collection, recycling, reuse, trade, and disposal. In addition, it accounts for the relevant material stocks in tailings; products in use, and landfills; (see the complete list of flows in Supporting Information S1). A recent MSA study covered five raw materials used in batteries, namely Co, Li, Mn, Ni, and NG (Matos et al., 2020). The manuscript series discussing the related outcomes of this work consists of four papers, three of which focus on the individual raw materials complete MSA (Ciacci et al., 2021; Godoy León et al., 2021; Lundhaug et al., 2021).

Here in the current study, a multilayer system approach (represented in Figure 2) is applied to the five MSAs using LIBs as the driving link between them. This was modeled while taking into account the same datasets and assumptions and then refined further (explained in Section 2.4 and Supporting Information S1) for all of the individual raw material cycles. By using this approach, the authors ensured that the mass balances are respected through all the layers of the system and mass balance is withheld across several dimensions, that is, LIBs and individual raw materials cycles.

Figure 2 is a schematic representation of the model developed and the layers studied for the example of Li in portable batteries. The model has a "grandparent" layer representing the total demand for all LIB applications in the EU-28. This "grandparent" layer has four "parent" layers (schematically represented in Figure 2 by layer (b)), each representing an individual LIB application: (1) mobility, (2) portable, (3) industry, and (4) e-bikes. Each of these layers has five "child" layers, one for each battery raw material (schematically represented in Figure 2 by layer (c)). The "parent" layer drives the five "child" layers through the demand for LIBs in the individual applications. In total, the LIB system under study was modeled using four battery application cycle layers and 20 raw materials cycles layers (one for each of the target materials in the four battery applications considered). In turn, each of these raw material life cycle layers is a fragment of the complete individual MSA, where all the applications are computed (Ciacci et al., 2021; Godoy León et al., 2021; Lundhaug et al., 2021). In the current publication, the flows of each material in the EU Li-battery system (represented in Figure 2 by layer (d)) are compared with the MSA flows of all applications within the same system boundaries (represented in Figure 2 by layer (e)).

This approach facilitates the study of the interconnections between individual raw materials and the supply chain in which they play a vital role, which can provide insights into potential critical bottlenecks for the individual materials as well as in the supply chains they are part of. The data structure for the use phase follows the ProSum dataset (see Section 2.4) and takes into account average battery lifetimes to calculate trends of the battery stocks and related waste generation trends. This allowed a detailed differentiation between battery applications of portable, industrial, and mobility batteries.

Song et al. (2019) also made a similar analysis for the same set of raw materials with a focus on the LIB sector in China. Their model includes an analysis of the criticality of the materials as well as the upstream flows of the LIB sector, at extraction and processing stages. In the present study, these stages are analyzed in the individual raw material publications (Ciacci et al., 2021; Godoy León et al., 2021; Lundhaug et al., 2021). In addition, the current work includes an analysis of the raw material used in other sectors so as to frame the LIB system within the complete EU market for the targeted materials (represented by layer (e) in Figure 2). This has several advantages as described in Section 3. Song et al. (2019) also modeled the evolution of the system until 2025, which could be a future improvement on the current system.
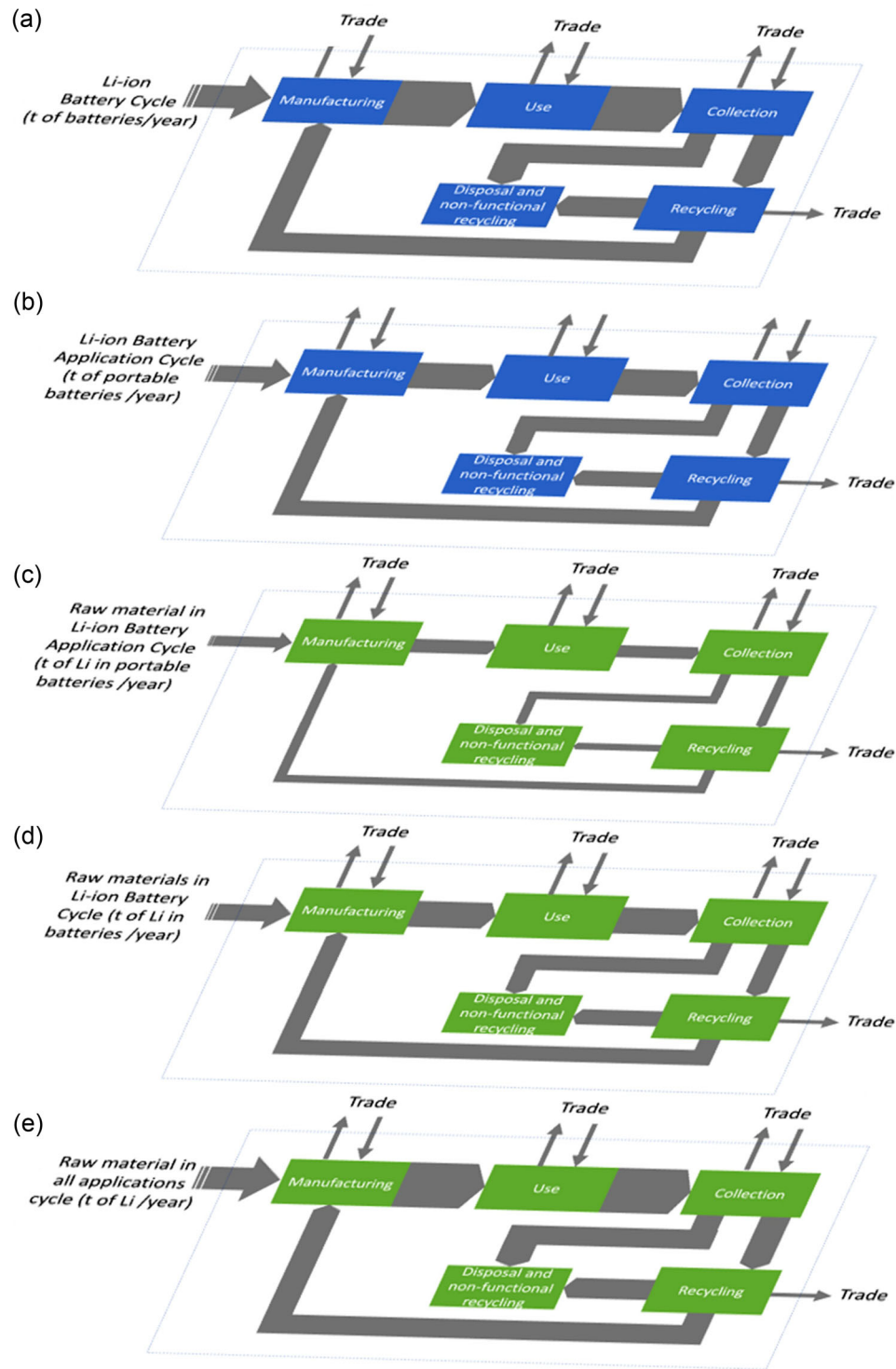
FIGURE 2 Schematic representation of the multilevel systems approach. In general, the approach models several cycles in each layer, namely: (a) "Grandparent" layer: One cycle translating the sum of all LIB cycles; (b) "Parent" layers: Four cycles of the single application LIB cycle (portable, mobility, e-bikes, and industrial batteries); (c) "Child" 20 layers: Cycles of raw materials (Co, Li, Mn, NG, Ni) in each battery application; (d) Five cycles translating the sum of the raw material in all LIBs; (e) Five cycles translating the sum of the raw material cycles in all applications. There is a decrease in mass from (a) to (c) and an increase from (c) to (e)

2.3 | Performance indicators

Six performance indicators described in Table 1 were selected to assess the efficiency of the EU LIB and raw materials system in terms of the EU: (i) self-sufficiency for supplying raw materials to the LIB sector with the self-sufficiency potential (SSP) indicator, (ii) recycling capability with three indicators: end-of-life recycling rate (EoL–RR), recycling process efficiency rate (RPER), total scrap recycling input rate (TS–RIR), (iii) accumulation of raw materials in the use phase with the in-use accumulation rate (IUAR) indicator, and (iv) raw materials losses after use through waste streams indicated by the post-consumer waste rate (PC–WR). The recycling indicators were obtained from UNEP (2011) and Tercero Espinoza and Soulier (2018), while the updated recycling rates data were extracted from the individual papers (see Supporting Information S1 for more information). The SSP, IUAR, and PC-WR were developed to characterize the results of this study.

2.4 | Data sources and data gaps

Relevant aspects of the data for key life cycle stages are discussed subsequently.

2.4.1 | The manufacturing phase

The manufacturing of LIBs for the four battery applications defined in Section 2.1 is schematically represented in Supporting Information S1. However, it was not possible to characterize the manufacturing stage in the detailed form presented there, using the available statistics as data are lacking for several of the LIB manufacturing steps. The available data only allow the estimation of the volumes of batteries assembled in the EU and the trade balance of finished batteries and finished products. The modeling of LIBs manufactured and traded within the EU required several assumptions and estimates based on the available EUROSTAT ("Easy Comext," 2020) and UN COMTRADE statistics ("UN Comtrade | International Trade Statistics Database," 2020) (further details in Supporting Information S1). Consequently, the values obtained may be subject to great uncertainty. It was assumed that the production of Li-ion portable and industrial batteries in the EU was negligible in comparison to the production of mobility batteries. Furthermore, an average distribution of the main mobility batteries produced in the EU in 2016, was assumed to be: 60% NMC 111, 30% NMC 433, and 10% NCA, based on global market figures (Darton Commodities, 2018; International Energy Agency, 2019). The detailed compositions defined for these chemistries are described in Supporting Information S1.

2.4.2 | The use phase

The shares of uses of each of the materials targeted were obtained as a result of complete system analysis of those materials, performed by the authors and described in detail in the three related publications (Ciacci et al., 2021; Godoy León et al., 2021; Lundhaug et al., 2021). The use phase inflow, stocks, and outflow were directly obtained from an updated database of the H2020 ProSUM project (Huisman et al., 2017, 2020; RMIS, 2019). The database integrates several data sources to get a comprehensive picture of the overall battery flows within the EU market, including volumes of rechargeable batteries from Avicenne, Eurostat, Alternative Fuels Observatory, statistics from member states, and ProSUM data batteries contained in electronic equipment. It was further assumed that 20% of batteries leaving the stocks was exported from the EU for reuse: this is a typical ratio of export for reuse in electronic equipment, as highlighted by the ProSUM project.

2.4.3 | The collection phase

Collection was defined according to the following efficiencies obtained from the literature: 95% of mobility, 90% of industrial, 50% of e-bike, and 45% of portable batteries are collected for recycling (Recharge, 2018). The remainder was assumed to go directly to landfill or energy recovery. According to some sources, 3.5% of the portable batteries collected at end-of-life were exported, and this data apply mainly to batteries contained in WEEE (BAN, 2018).

2.4.4 | The recycling phase

Recycling efficiencies depend on the recycling technologies used, which have been evolving rapidly in recent years. Some technologies under development can achieve efficiencies of up to 98% for some electrode materials, but they were not all in place in the target year of 2016. Up to 50% of the battery weight may be lost in pyrometallurgical recycling processes due to thermal oxidation of metals, electrolytes, and plastics. Efficiencies

TABLE 1 Description and equations of performance indicators

Indicator	Acronym	Brief description	Formula
Self-sufficiency potential	SSP	It measures the amount of primary and secondary material input domestically supplied over the total inflow to use	$(B1.1^* + B1.2^* + G1.1^* + G1.2^*) \times \lambda / M3.1$ Where λ is the share of the raw material demand for LIBs
Total scrap recycling input rate	TS–RIR	It measures the fraction of total scrap from recycling of LIBs over the total material input to the manufacturing stage	$(G1.1 + G1.2 + D1.5 + C.1.4 + D1.9 + D.1.10) / (M.1.2)$
End-of-life recycling rate	EoL–RR	It measures the fraction of old scrap recycled over the total amount of old scrap generated in a given year	$(G1.1 + G1.2 + G1.3) / M4$
Recycling process efficiency rate	RPER	It measures the fraction of total recycled scrap (i.e., new scrap + old scrap) over the total scrap input into recycling in a given year	$(G1.1 + G1.2 + G1.3 + D1.5 + D.1.10) / (F1.4 + D1.5 + D.1.10)$
In-use accumulation rate	IUAR	It measures the fraction of the material stored in stocks over the total material placed on the use market in a given year	$(E.1.8 + E.1.7) / M3.1$
Postconsumer waste rate	PC–WR	It measures the fraction of total input to the use phase material loss over the total inflow to use in a given year	$(E.1.5 + F.1.3 + G.1.5) / M3.1$

Note: Flows used from the complete MSA of the raw material are represented by an (*). List of flows is represented in Supporting Information S1.
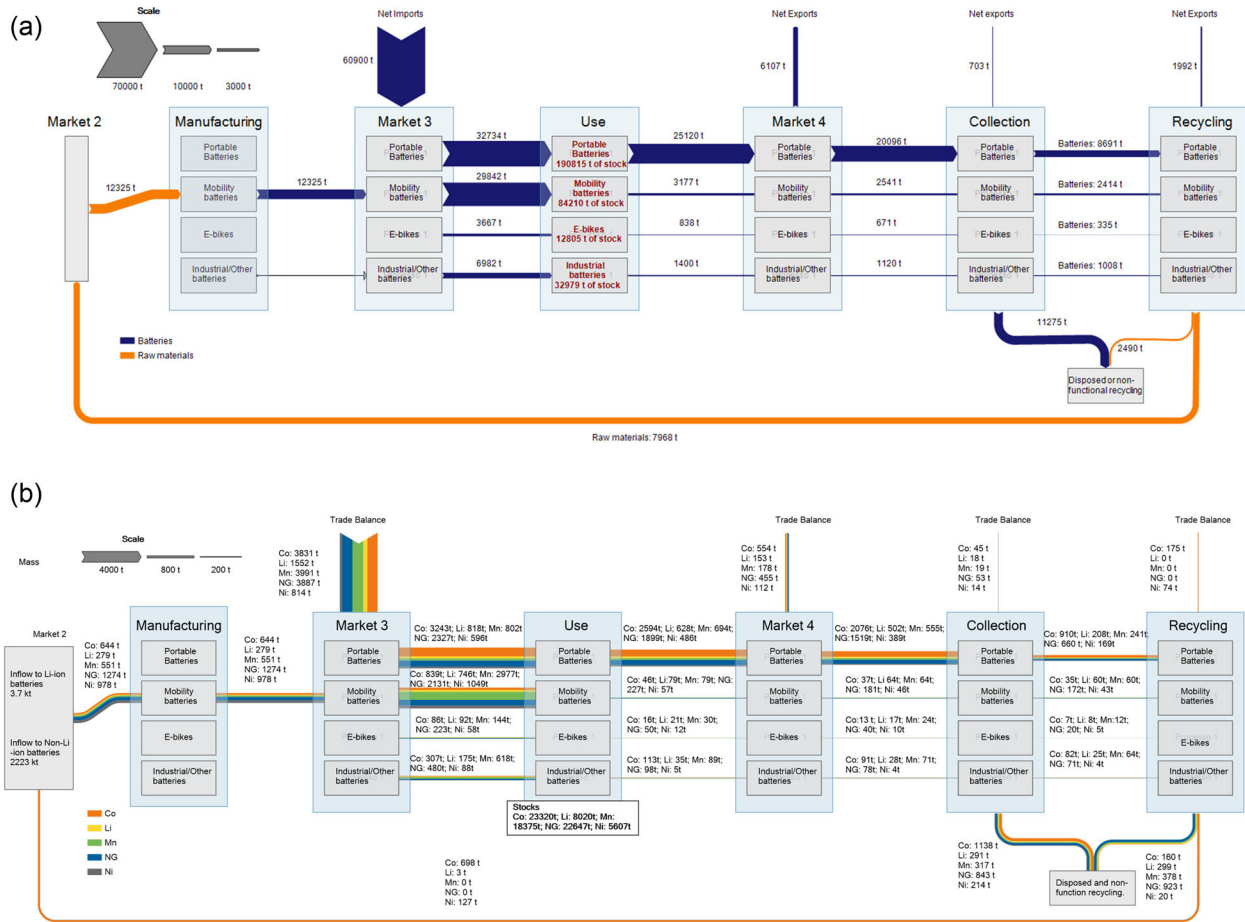
FIGURE 3 Flows of the (a) whole Li-ion battery cycles including cycles for the different battery applications (capturing layers (a) and (b) of Figure 2) (b) raw material flows within each Li-ion battery application (captures layers c) and (d) of Figure 2) (see Section 6 in Supporting Information S1 with the data and assumptions used to produce this figure)

greater than 80% may potentially be obtained by using hydrometallurgical processes (Harper et al., 2012; Kushnir, 2015). About 80% was assumed as an overall raw materials recovery efficiency of the recycling process for the whole battery system (Figure 2 layer (a)) while for the individual raw material cycles (Figure 2 layer (c)), the recycling efficiency was based on the literature and defined as follows: (a) for Co 85% and 80% (for portable batteries and for the other battery applications, respectively) (Kushnir, 2015; Sommer et al., 2015); (b) 1% for Li; (c) 0% for Mn, although state-of-the-art combined pyrometallurgical and hydrometallurgical recovery processes can currently recover Mn (as sulfate) from slag with efficiencies as high as 86% (Bobba et al., 2020; Recharge, 2018); these technologies were not widely implemented by the industry in 2016 (Cusenza et al., 2019; Lebedeva et al., 2017); (d) 0% for NG (Bobba et al., 2020; Recharge, 2018); and (e) 91% for Ni, (Bobba et al., 2020; Recharge, 2018).

3 | RESULTS AND DISCUSSION

3.1 | Stocks and flows in the multilayer system

3.1.1 | The EU Li-ion battery system: Applications and raw materials stock and flows

This section presents the detailed results of the flows and stocks of five battery raw materials in the EU LIB system for 2016, and the links of this battery system with the complete value chains of each raw material as described in three related publications by the authors (Ciacci et al., 2021; Godoy León et al., 2021; Lundhaug et al., 2021).

Figure 3a depicts the EU LIB cycles divided into four battery applications, they correspond to "grandparent" and "parent" layers described in Section 2.2 (layers (a) and (b) in Figure 2). Figure 3b demonstrates how the individual LIB flows of Co, Li, Mn, Ni, and NG are interconnected in the EU system, corresponding to the "child" layers described in Section 2.2 (layers (c) and (d) in Figure 2).

In 2016, the EU production of LIBs was rather limited, in total around 12.3 kt of LIBs were produced in the EU. This resulted in rather low raw material flows leaving manufacturing embedded in mobility batteries (see Figure 3b). This illustrates the lack of LIB manufacturing capacity in the EU, a well-known bottleneck to achieve competitiveness in this sector: The EU production represented only 3% of the global Li-ion cell manufacturing (Lebedeva et al., 2017). What may be not so well reported are the flows of the different critical raw materials entering the EU embedded in LIBs and finished products, as reported in Figure 3b as the trade flow entering the market 3 stage. This information may be crucial to understanding upstream supply chains of those batteries better and supporting future solutions to alleviate EU dependency by possible internal supply of some of the raw materials required. In 2016, net imports of 60.9 kt of LIBs and their products (e.g., electrical vehicles, tables, e-bikes) entered the EU, which represents more than 83% of total EU demand. These imported products mainly contained Co, Mn, and NG; the shares of Li and Ni were low in 2016, because Ni-enriched chemistries were not dominant in the use phase. Even though Li is the core component of the battery's electrochemistry, its concentration in the whole battery is low (2.5% on average).

In general, in 2016, a total of 73.2 kt of LIBs were placed on the EU market and this amount was mainly divided between portable batteries and mobility batteries. E-bikes and industrial batteries represented a smaller share of the EU market. Figure 3b shows that Li was present in smaller quantities in the use phase due to its low concentration in LIBs. From the raw materials analyzed, Co was the principal material in terms of volume contained in portable batteries placed on the market due to its use in LCO and NMC chemistries. Mn dominated the chemistry of mobility batteries due to its use in LMO and NMC batteries. The EU imports LMO batteries mainly for their portable and mobility battery needs. NG was the second main component of the electrodes of the LIBs used in the EU, constituting 7% of the whole batteries, on average. Compared to its main applications, Ni was used in modest quantities in 2016 (maximum 5% of the whole battery on average). However, Ni use in LIBs has been increasing gradually over the years as it is expected to partially substitute Co in new battery chemistries (due to Co supply risk problems and price, NCM 622 and NCM 523 have been replacing the equally balanced NCM 111 chemistry; Alves Dias et al., 2018). Ni cannot, however, substitute Co entirely and will always be used in combination with it.

Regarding stocks in use, in 2016, there were 321 kt of LIBs in use or hibernating (i.e., kept by the users after end-of-life). Portable batteries represent 59% of these stocks while the other battery applications analyzed represent between 4% and 26% of the total stock. This is not only explained by higher volumes of portable batteries being put on the market every year, but also because, in contrast to mobility batteries, they have been available on the market since the 90s and are often kept by the users at end-of-life due to their small sizes. On the other hand, mobility batteries (except for e-bikes) were marketed after 2010, but their demand is increasing fast and the stock accumulation situation has increased gradually (see Supporting Information S1 for more information on the gradual evolution of the stocks). In addition, the majority of the EU in-use stock is built using NMC and LCO battery chemistries and this explains the results achieved for the individual battery materials showing a dominant share of Co, Mn, and NG. It is important to highlight this since the quantification of the volumes and composition of this stock is crucial in defining future resource recovery strategies through recycling or reuse of batteries (Bobba et al., 2019). Therefore, the existence of such stock potentiates solutions to alleviate EU dependence on LIBs and its raw materials imports once the in-use batteries become available for collection and treatment. This is actually why the European Commission has proposed ambitious targets concerning collection, recycling efficiency, recovery of materials, and recycled content in its proposal of an EU regulation on batteries and waste batteries (European Commission, 2020c). This is also why European member states have been massively investing through the ICPEI schemes, into pan-European research and innovation projects along the entire battery value chain that fully integrate the circular economy dimension. Collection and recycling may be faster for portable batteries that have lifespans between 2 and 7 years (depending on the application) (Godoy León & Dewulf, 2020) than for mobility batteries with lifespans up to 10 years (Eurobat, 2018). Additionally, to optimize the earlier availability of batteries for reuse or recycling, the time that these products are kept by the users after their life time should be minimal. Nigl et al. (2020), for example, reported considerable amounts of portable batteries hoarded in European households. The situation for mobility batteries may be different as the hoarding time tends to be close to zero although limited information exists on this topic.

Nearly 12.5 kt of batteries were collected for recycling taking into account the collection efficiencies reported in Section 2.4. As reported by Nigl et al. (2020), considerable amounts of portable batteries are either entering residual waste collection streams or misplaced into nonbattery-specific collection systems. In the case of mobility batteries, collection rates are higher than portable batteries due to: (i) lower multiplicity of end products which facilitates collection; (ii) existence of specific legislation on end-of-life vehicles (European Commission, 2000, 2005); (iii) longer lifetimes and later penetration on the market which means that the in-use stock is still building up and there is still sufficient capacity to treat mobility batteries leaving the use phase; (iv) large sizes that prevent storage at home and inappropriate collection, for example, in municipal waste streams. Analyzing the raw materials embedded in the batteries, their collection efficiencies reduced the capacity of the LIB system to achieve circularity by around half. Such a result clearly indicates the need to increase efforts on battery collection through specific policy provisions.

The EU LIB system was able to recycle 0.7 kt of Co and 0.1 kt of Ni and less than 0.03 kt of Li, Mn, and NG. This is a consequence of the high recovery rates achieved for Co and Ni and low-to-nonexistent recovery of Li, Mn, and NG in 2016. These three materials either ended in landfill or recycled as tramp elements into other material cycles, exiting the LIB system (i.e., nonfunctional recycling). Such losses prevent circularity in material cycles and with it the potential for recycling to mitigate supply security issues and to significantly improve resource and energy efficiency and the environmental impacts of the EU's LIB supply chain (Cusenza et al., 2019; Simon & Weil, 2013; Unterreiner et al., 2016). Reuse/repurposing
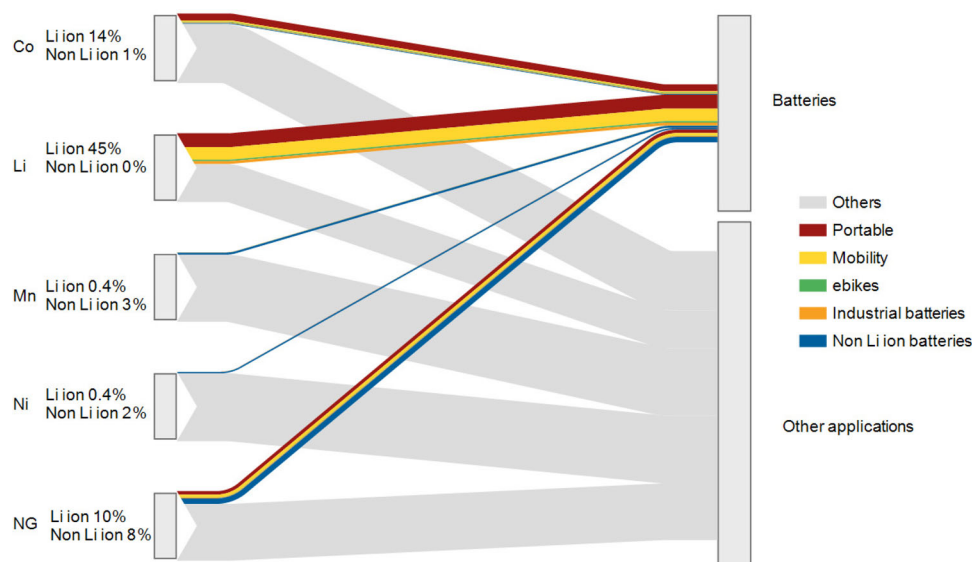
FIGURE 4   Share of end uses between battery applications and other sectors for the five battery raw materials under study, in the EU for the year 2016. Note: These shares translate the uses of cobalt in the EU and in 2016; the current share of Co used in LIBs is now higher than 14%, in the EU. Additionally, globally, the cobalt main application is in the production of batteries, particularly in LIBs (see Section 7 in Supporting Information S1 with the data used to produce this figure)

of LIBs is another alternative at end-of-life, as yet not widely diffused in the EU but is gradually being applied in a few countries (Bobba et al., 2019; Liu et al., 2021).

Since 2020 the United Kingdom (UK) is no longer part of the EU, an overall reduction in the amount of raw materials and LIBs entering the use phase in the EU is expected. However, no substantial differences were observed except in the cases where the UK is a main producer of the raw material as that is the case for nickel metal. In this case, the departure of the UK from the EU will increase the dependency of the EU on imports of nickel and nickel products.

The demand for LIBs in the EU has recently been changing significantly (Huisman et al., 2020); in Supporting Information S1, a time series of evolution of the LIBs demand and stocks is presented.

One of the main driving forces for the development of new recycling technologies and routes is the scarcity of key raw materials, price of raw materials, and their problems of supply. The paper clearly demonstrates the interconnections between raw materials along the battery value chain, considering individual materials but also primary and secondary raw materials. It provides also quantitative estimates about total in-use stocks and annual material flows generated at end-of-life. This is essential information for the planning of recovery strategies and it may ultimately lay the basis for economic feasibility of a reliable and long-term sustainable recycling industry in the EU for critical or expensive raw materials. For example, cobalt is an expensive, increasingly rare raw material with significant supply issues. The recycling of LIB chemistries containing Cobalt may potentiate the recovery of other less expensive raw materials, such as Ni and Mn, present in the same chemistries. In addition, the increasing dependency of battery grade Li carbonate may not only push the development of economically viable recycling technologies for Li recovery but also of new mining projects dedicated to the extraction of Li suitable for this grade. The information of these interconnections is also important for the setting of material recovery strategies at end-of-life. For instance, the recovery of raw materials from battery chemistries containing Mn and different percentages of Li and Co will require combined pyrometallurgical and hydrometallurgical recovery processes for Mn recovery.

3.1.2 | The EU Li-ion battery system: Comparison with other applications

The link between the results for the EU LIB system and the individual materials value chains is translated in Figures 4 and 5 (which aggregates results from layers (c), (d) and (e)). These figures represent the shares of each raw material in LIBs and in other applications sent to use, stocks, disposed of, and functionally recycled. The results on the other applications are detailed in three related publications on the life cycles of the individual materials (Ciacci et al., 2021; Godoy León et al., 2021; Lundhaug et al., 2021).

With regard to the availability of battery raw materials in the EU battery value chain, it is important to highlight that LIBs for the majority of the targeted raw materials are not the main application in use in the EU in 2016, with the exception of Li that currently has 45% of its end uses directed to batteries. LIBs have to compete with a wide variety of applications, such as glass and ceramics, superalloys, refractories, and building
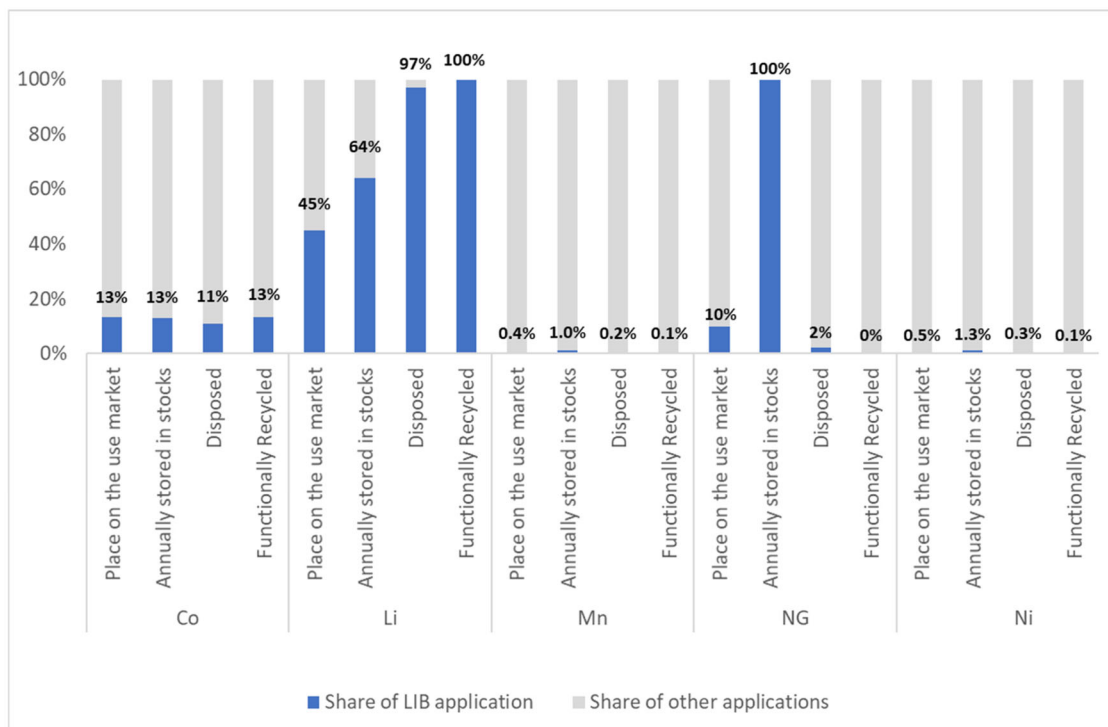
FIGURE 5 Share LIBs (blue) and other applications (grey) contribution to the individual material flows for the following flows: Inflow to use (place on the use market), annually stored in stocks, disposed, and leaving functional recycling (including all applications and end-products) *Note*: Annually stored in stocks refers to the share of material in LIBs (blue) and other applications (grey) that is annually added to in-use stocks. LIBs represent the only source of secondary Li in the EU (therefore, LIBs represent 100% of the flow of recycled material) in 2016 (see Section 8 in Supporting Information S1 with the data used to produce this figure)

and construction, for the availability of raw materials. This last fact may, however, change in the near future if demand for batteries continues to grow. This could be a major bottleneck in achieving the desired increase in LIB manufacturing capacity of the EU, which could only be detected by the multilayer analysis proposed (see Figure 2). This new result appears to be extremely important for policymakers in ensuring a sustainable supply of battery raw materials in the EU as planned in the EU Strategic Action Plan for Batteries (European Commission, 2018b). The expected increase in demand for LIBs (European Commission, 2020a) may change the representativeness of these batteries in the raw materials life cycles, especially Ni and NG where batteries now represent a smaller subsection of their supply.

The main volumes of Co going annually into stocks are embedded in superalloys, which have higher lifespans and are the main end uses for Co (see Figure 5 and Co MSA publication for more details; Godoy León et al., 2021). On the other hand, batteries are the main products that store Li and NG annually in stocks since the other applications are either dissipative or their stocks are decreasing due to reduction in consumption (for the case of NG only LIBs contributed positively to stocks increase in 2016). LIBs constitute less than 1.3% of Mn and Ni entering the in-use stock annually, building and construction, and engineering sectors are at the top of the list of sectors storing them, which have high lifespans and are also the main applications of Mn and Ni (see Figure 5 and Co MSA publication for more details; Ciacci et al., 2021).

Overall, the volumes of Co, Mn, and Ni recovered from LIBs were marginal compared to the total amount recovered from all main end-uses in the EU. Current technologies are only able to recover 1% of the Li in LIBs, but this still represented the only source of secondary Li in the EU (therefore, LIBs represent 100% of the flow of recycled material; see Figure 5) in 2016. No functional recycling existed for NG. Comparing the recycling of LIBs from other applications such as superalloys, the latter are recycled to a great extent with the recovery of materials, such as Co, at rates between 80% and 90%.

One key result of this analysis is that it demonstrates that the majority of the nonbattery applications of Li are dissipative (except Li in glass and ceramics that is stocked in use but not recovered in the recycling stage). Therefore, the efforts to secure internal supply for this material to the EU should essentially come from improvements in collection and recycling of batteries and the development of domestic extraction (to the extent domestic reserves allow). This also justifies the current proposal of policy incentives to increase recycling efficiency of key raw materials, in particular for Co, Ni, Li, from LIBs in the proposal for an EU regulation on batteries and waste batteries. The quality of recycled material will also be essential to alleviate EU dependence on imports, as reported by Ziemann et al. (2018).
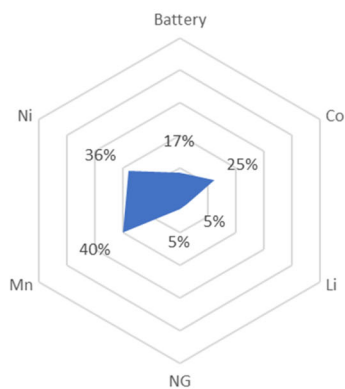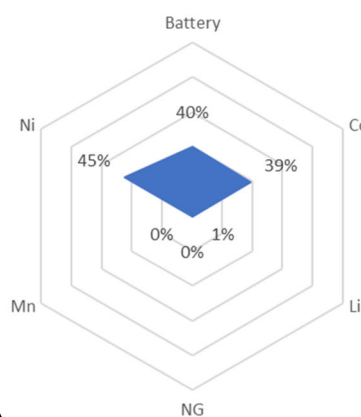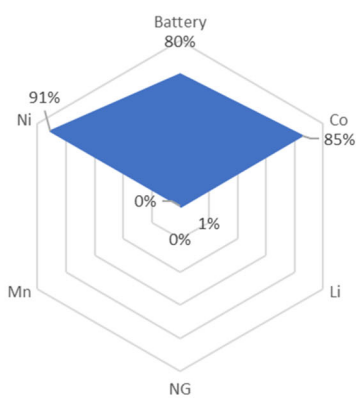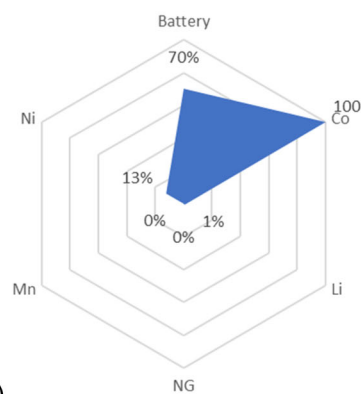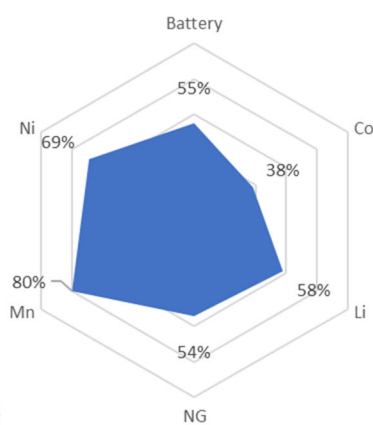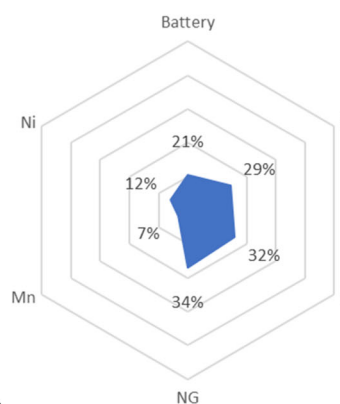
FIGURE 6 Indicators for whole battery and single raw materials only considering the battery flows in the EU in 2016 (see Section 9 in Supporting Information S1 with the data used to produce this figure)

3.2 | Analysis of indicators

The EU is dependent on imports of LIBs, their components (e.g., battery cells and their final products), and raw materials required for their manufacture. Even if the EU is able to supply more than 25% of its demand for Co, Mn, and Ni from domestic sources, the LIB market requires these materials to be battery grade, which requires further processing of the materials. Therefore, EU self-sufficiency able to supply raw materials with appropriate characteristics to the EU battery market would probably be significantly lower than the shares in Figure 6a. The situation is different

regarding Li and NG because there is currently no production of battery grade Li and NG in the EU. Almost all spherical graphite was produced in China. Furthermore, no installations for the production of cathode materials operated in the EU at the end of 2018, whereas in 2020, the EU cathode production amounted to only 500 tonnes. The lack or limited capacity of the EU to supply battery grade materials from the volumes extracted domestically clearly represents a bottleneck to the EU building up LIB manufacturing capacity. In 2016, this capacity only represented 17% of the battery demand in the EU (see Figure 6a). However, there are several noticeable efforts in the EU to reduce the dependency on battery grade raw materials (European Commission, 2020c; Terrafame Oy, 2018; Terrafame, 2018).

The share of material recovered through recycling after collection of LIBs is close to 40% for Co and 45% for Ni (see Figure 6b); the rest is lost or exported due to: (i) inefficiencies in collection and (ii) exports at end-of-life. As explained earlier, the recycling of Li is still under technological development while the NG and Mn collected in 2016 were not recovered. This represents an important obstacle to increasing the circularity of materials in the EU LIB system. The great discrepancies of recycling performances between battery raw materials might justify policy actions, including potential provisions on material recycling efficiencies for specific materials of LIBs and on content of postconsumer recycled materials, as currently proposed in the new EU regulation on batteries and waste batteries.

Figure 6d provides a measurement of circularity of the EU LIB. This rate is remarkably low for all the materials except for Co. The value estimated indicates that the demand of Co for LIBs manufacturing in 2016, could be fulfilled by the amount recovered in recycling of LIBs. However, this evidence has to be carefully interpreted due to the great uncertainty of the estimates of the EU battery manufacturing capability and of disputable assumptions about the characteristics of battery grade materials. Additionally, the scrap obtained from the EU LIB system currently represents less than 2% of the total input of this material to the EU for all applications. If the same volume of recycled Co is compared with the input of Co to the use phase (i.e., combining EU production and imports) of the LIB system, then a rate of 31% is achieved (see Market 3 of Figure 3b).

The accumulation (see Figure 6e) and postconsumer loss rate (see Figure 6f) show that the majority of the materials accumulate in use or in hibernating stocks. The exception is Co that mainly leaves the use phase in the form of portable batteries and is then either exported, wasted, or recycled in larger volumes than the other materials. Li and NG have the highest shares of loss through waste streams in comparison with other materials (see Figure 6f) because of their minor recycling rate and null nonfunctional recycling. The low loss rate presented by Mn (Figure 6f) is mainly explained by the nonfunctional recycling of this material and great accumulation in the use phase in 2016.

4 | CONCLUSIONS

The flows and stocks of LIBs and five of their raw materials (Co, Li, Mn, NG, and Ni) within the EU were determined using a multilayer system approach, which visualized significant relationships across two dimensions: LIB applications cycle and raw material cycles.

It was clearly demonstrated that a coordinated strategy addressing extraction, manufacturing, collection, and recycling needs to be developed by the EU to enhance its manufacturing capacity and competitiveness in the global LIB market. It was shown that individual materials face different challenges that need to be solved by also considering the interdependencies with battery and other raw materials cycles in order to avoid potential problem shifting. While internal EU refining capacity for battery grade materials exists for Co, Ni, and Mn, this is not the case for Li and NG where all battery grade refining occurs outside the EU. This means that the supply of Li and NG could become even more critical and this represents one of the main bottlenecks to achieve the desired increase in manufacturing capacity for LIBs. The ongoing gradual substitution of Co with Ni in the battery chemistries might disrupt the EU Ni cycle particularly for its use in less valuable applications with Ni potentially to be diverted to LIBs. In addition, Co is usually a byproduct of Ni extraction and developments in the LIBs chemistry might also influence the upstream flows of both raw materials, and this may increase the need for the construction of new refining facilities in the EU.

The current work provides novel insights into sustainable resource management of LIBs and their raw materials for EU policies, namely: (i) interconnections between different materials in all the lifecycle stages; (ii) quantification of the competition for raw materials using other applications detailed in the individual raw materials MSA; LIBs are not currently the main applications of the raw materials targeted in this study (except for Li); (iii) importance of developing efficient collection strategies of LIBs, particularly for portable batteries, which presented lower collection rates and high accumulation in hibernating in-use stocks; (iv) importance of developing targeted recycling strategies for Li and NG as the other applications of these materials are essentially dissipative and the recycling infrastructure for these materials still needs to be built to achieve cost-effective recycling; (v) information on composition of stocks in terms of LIBs, their raw materials, and relationships with other application products, important in inferring availability of resources at end-of-life; and (vi) information on composition of the trade flows is also key for trade agreements, for example.

Future work to increase the relevance of this approach may rely on its use in combination with social and environmental assessments aimed at improving sustainable sourcing in the EU LIB system. A systematic analysis of the system time-series can help in creating predictive models that define the future relationships of the battery raw materials studied. Finally, the same multilayer approach can be applied to other sectors such as space technologies.

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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