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Technical, economic and environmental analysis of production scraps direct recycling from lithium-ion battery manufacturing

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ABSTRACT

To address the well-known limitations of current recycling methods, particularly the challenges associated with heterogeneous and degraded end-of-life (EoL) lithium-ion batteries (LIBs), including complex disassembly, electrolyte removal, and material cross-contamination, this study proposes a practical and efficient alternative based on direct recycling of production scraps. In this study a direct recycling process, which recovers active materials without altering their crystal structure, consisting in thermal treatment followed by mechanical detachment through ball milling, was applied to production scraps of lithium iron phosphate (LFP) and lithium nickel manganese cobalt oxide (NMC) cathodes. Different temperatures (200-300-400-600 °C) and air or nitrogen were explored. Experiments showed that partial melting of PVDF binder at 200 °C was sufficient to enable effective material recovery, particularly for LFP, without inducing degradation. The highest recovery yields were recorded at 400 °C in air for LFP (98 ± 7 % Fe and 99 ± 11 % Li) and at 600 °C in air for NMC (99 ± 19 % Co, 99 ± 12 % Li, 99 ± 17 % Mn and 99 ± 5 % Ni). Thermal treatment at 300 °C led to PVDF melting without degradation which increased the concentration of Al impurities in both LFP and NMC materials, likely due to the binder's molten state enhancing adhesion to the Al foil prior to its complete thermal decomposition. The economic viability of the process was confirmed by recovery costs (0.71 €/kg LFP and 1.17 €/kg NMC) lower than current virgin material prices. Greenhouse gas (GHG) emissions equal to 1.18–2.56 kg CO_2/kg for LFP and 1.94–6.98 kg CO_2/kg for NMC were calculated, with the most favourable trade-offs achieved at moderate temperatures in air. Due to low energy demand and easy scalability, the proposed direct recycling process holds potential for on-site recycling of scrap cathodes, particularly in regions -such as the European Union, where critical raw materials supply security and waste reduction are significant issues.

1. Introduction

The rapid growth of the lithium-ion battery (LIBs) market, driven by global efforts to decarbonize the transportation and energy sectors, is expected to increase LIBs demand from 700 GWh in 2022 to 4700 GWh by 2030 (Statista, 2024). This will have significant consequences on the supply chain for LIBs raw materials, particularly in terms of sourcing security, market stability, and the environmental impact of mining activities (Mayyas et al., 2019; Vinayak et al., 2024). Critical raw materials as Co, Li, Ni and graphite are essential for producing LIBs, yet they are geographically concentrated and limited in availability, raising both physical scarcity and supply risk concerns (Olivetti et al., 2017). In this context, recycling spent LIBs to recover secondary raw materials can stabilize supply chains while reducing waste from end-of-life (EoL) LIBs (Lv et al., 2018; Miao et al., 2023).

Direct recycling strategies are particularly promising, as they not only recover valuable materials but also aim to reintegrate them directly into new cells (Zhou et al., 2024). State-of-the-art recycling methods as pyrometallurgy and hydrometallurgy rely on high-energy demand long-loop processes to individually recover metals or mineral precursors for the manufacturing of new battery cells. On the other hand, direct recycling retains and restores the structure of active materials in the electrodes, thereby minimizing both energy use and environmental impact (Ma et al., 2025; Xu et al., 2023). Direct recycling has

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demonstrated strong potential for both material recovery and performance restoration (Cao et al., 2024), and can reduce energy consumption by up to 80–90 % and emissions by approximately 60 % compared to state-of-the-art recycling processes (Xu et al., 2020). Nonetheless, direct recycling technologies face several challenges, such as the need of labour-intensive disassembly and sorting (Ji et al., 2023), the lack of standardization in battery designs (Rehman et al., 2025), and scalability, with loss of efficiency due to the scale up of the process (Roy et al., 2024). Sorting accuracy is a critical issue in direct recycling, as recycling processes are typically chemistry-specific, yet many batteries lack clear labelling and existing sorting technologies are not yet fully developed (Wei et al., 2023).

In contrast to heterogeneous and degraded EoL batteries, production scraps represent a pristine, structurally intact, and chemically uniform feedstock, making them an ideal candidate for simplified direct recycling processes that bypass the complexities of sorting, discharging, and disassembly (Sita et al., 2024). Production scraps derive from LIBs manufacturing (e.g., electrode trimmings and components that fail quality checks), and in a smaller amount from the research and development stage of new battery technologies (Yu et al., 2024). As LIBs manufacturing scales up in the next future, also the amount of production scrap material will grow significantly (Gaines et al., 2021). With an average generation rate between 5 % and 30 % of scraps (Gaines et al., 2023), it is important to note that the production of scrap is more significant at the beginning of operations. Studies report that scrap material can account for as much as 28.8 % of production after the first year, with this figure decreasing to 8.6 % after five years (Dahamen et al., 2024). Production scraps are pristine, structurally intact, and uniform in chemical composition and physical structure, making them ideal for efficient direct recycling (Wei et al., 2023). Therefore, direct recycling processes applied to LIBs production scraps can be simplified into short-loop approaches focused on recovering active materials through detachment from current collectors without compromising their electrochemical performance.

A key critical issue for direct recycling of production scraps is Polyvinylidene fluoride (PVDF) binder, which is used to ensure material adhesion to the current collectors (Gupta et al., 2024). Common detachment methods include chemical (Song et al., 2014) and thermal processes (Gao et al., 2024). Chemical detachment methods involve the use of solvents to dissolve the PVDF binder, such as N-methyl-2--pyrrolidone (NMP), with efficiency of 86.70 % after 50 min at 60 °C (Liu et al., 2020) or up to 99 % after 90 min at 70 °C applying an ultrasonic power of 240W (He et al., 2015). The use of NMP is limited due to its high volatility and toxicity, raising environmental and safety concerns (Sherwood et al., 2016). Alternative solvents, such as Dimethyl Sulfoxide (DMSO), glycerol triacetate or cyrene offer comparable properties to NMP, however, they still face challenges regarding scalability, safety, and economic feasibility (Bai et al., 2020a; Elmaataouy et al., 2023; Gercek et al., 2024; Wang et al., 2020). Whereas thermal treatments target the physical degradation of PVDF to loosen the bond between active material and current collectors (Kim et al., 2021). Over 90 % of active material can be recovered through thermal treatment at temperature above 500 °C in oxidizing (Jafari et al., 2020) or inert atmosphere (Zhang et al., 2018). Thermal processes are often presented as safer alternative to chemical processes because they don't require toxic solvents such as NMP; however, high-temperature decomposition of PVDF can also generate hazardous gaseous by-products (Hu et al., 2022; Lombardo et al., 2020). Therefore, optimizing thermal process conditions is recommended to minimize these risks while maintaining detachment efficiency (Bruno and Fiore, 2025).

Despite production scraps could potentially represent an important feedstock for direct recycling processes, most existing studies on detachment enhanced by thermal treatment focused mostly on EoL cathodes (Chen et al., 2024; Zhong et al., 2019). Besides, among the few studies that investigated thermal treatment of production scraps, the interest has been predominantly on Lithium Nickel Cobalt Manganese Oxides (NMC) cathodes (Hanisch et al., 2011; Lombardo et al., 2021; Zhang et al., 2016). In our previous work (Bruno and Fiore, 2024a), we examined detachment methods for recovering active materials from current collectors in both end-of-life Lithium Iron Phosphate (LFP) cathodes and production scraps, with results indicating lower recovery efficiency in LFP production scraps due to the pristine condition of the binders, which hinder detachment. However, a significant research gap remains in the lack of comparison between direct recycling methods for different cathode chemistries, such as LFP and NMC production scraps. This study uniquely addresses this gap by directly comparing the recovery efficiency, purity, and structural integrity of both cathode types under identical processing conditions, providing a quantitative evaluation of their relative performance. Direct recycling was explored through a process consisting of three simple steps: (i) thermal treatment, varying process temperatures (200-300-400-600 °C) in oxidative and inert (N_2) atmosphere, (ii) ball milling and (iii) sieving at 1 mm to separate the recovered active material from the Al current collector. The main goal of the study was to identify the optimal conditions to maximize recovery efficiency without compromising the purity and the integrity of the active cathodic materials. Energy consumption was directly measured during the experimental process, to assess the economic and environmental impacts of the proposed direct recycling processes.

2. Materials and methods

2.1. Materials and equipment

Production scraps of LFP and NMC cathodes were supplied by the National Research and Development Institute for Cryogenic and Isotopic Technologies – ICSI Rm. Valcea, in Romania. The production scrap samples consisted of 20%wt. of current collector (Al) and 80%wt. of active material, containing 3%wt. of C65 conductive carbon black, 5 % PVDF and 92%wt. of cathodic active powder. The active materials were manually detached from the current collectors with a spatula.

Direct recycling tests were performed using a Carbolite GERO TF1 12-60-300 tubular muffle furnace in air or nitrogen (2 L/min flow) at different temperatures between 200 and 600 °C applying a 5 °C/min heating rate and then keeping the target temperature for 30 min; a Retsch MM200 planetary ball mill with zirconium oxide milling chambers and beads, and a 1 mm Giuliani particle-size sieve.

The content of Al, Co, Fe, Li, Mn and Ni (mg/kg) in the samples was analyzed via Flame atomic absorption spectroscopy (FAAS) (Schimadzu, GFA-EX7) and X-ray fluorescence (XRF) spectrometry (Rigaku, NEX DE). Before FAAS the samples were digested in aqua regia in a MILESTONE ETHOS UP microwave digestor. The presence of PVDF and graphite was identified through Fourier-transform infrared (FTIR) spectrometry (Thermo Fisher Nicole Summit). The crystalline structure was recognized by X-ray diffraction (XRD) spectroscopy (PANalytical X'Pert) with a Cu Kα radiation.

2.2. Experimental procedure

The direct recycling process tested in this study on LFP and NMC scrap cathode samples consists in three steps (Fig. 1): (i) a thermal treatment of the scrap cathodes for 30 min, explored in different conditions by varying temperature and gas flow (200-300-400-600 °C in air and to 400–600 °C in N_2), (ii) ball milling for 5 min at 15 Hz, and (iii) sieving at 1 mm. All tests were performed in triplicates on 2 g samples.

The efficiency of the direct recycling process was calculated based on the separation efficiency η of individual metals (Fe, Co, Li, Ni and Mn) according to Eq. (1),where "$c_{i,1}$" and "$c_{i,2}$" are respectively the concentration (μg/g) of the ith element in the initial samples and in the recovered material while "m_1" and "m_2" are the mass (g) of the initial samples and the mass of recovered material.

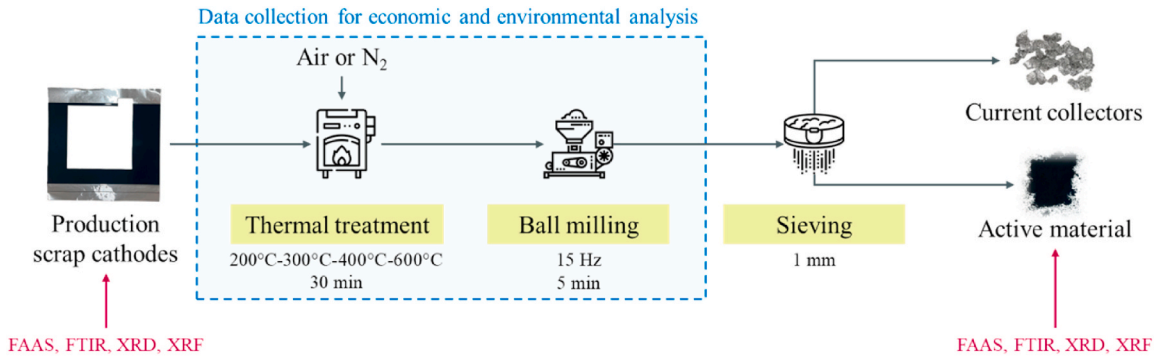

Fig. 1. Flowchart of experimental activities, operative conditions and analytical techniques applied in this study.

$$\eta_i(\%) = \frac{c_{i,2} \cdot m_2}{c_{i,1} \cdot m_1} \quad (1)$$

2.3. Economic and environmental analyses

A preliminary analysis of the economic and environmental impacts associated with the proposed direct recycling processes was conducted, adopting a "gate-to-gate" system boundary that includes the thermal treatment, ball milling, and sieving steps, and excluding the upstream impacts from the production of LIB cathodes generating the production scraps. Energy and materials (N_2 gas) consumption measured during the various steps of the experimental tests. Energy consumption was measured with a commercial powermeter (Maxcio PM10) for small laboratory batches and normalized considering the maximum treatment capacity of the specific equipment (e.g., 20 g for the Carbolite GERO TF1 furnace; 10 g for the Retsch MM200 ball mill). Energy consumption initially measured was extrapolated linearly to a functional unit of 1 kg of processed scrap based on the maximum effective batch capacity of each piece of equipment.

The analysis aimed to calculate the cost and CO_2 eq emissions related to the direct recycling of 1 kg of scrap cathodic material and to the recovery of 1 kg of active material to produce new battery cells, considering the separation efficiency and the purity of the recovered active material. The economic analysis was based on the separation efficiencies (see 2.2), on the average European market price of energy for non-household consumers, including taxes and levies, of 0.23 €/kWh (Eurostat, 2024) and N_2 gas (Ecoinvent, 2024), and on the average market price of LFP and NMC materials (Gaines et al., 2021; Manjong et al., 2024; Orangi et al., 2024). A sensitivity analysis was also carried out to calculate the threshold prices of electric energy and N_2 gas limiting the economic viability of the proposed direct recycling process. Finally, a preliminary environmental analysis was estimated the greenhouse gas (GHG) emissions associated with the direct recycling process and related to energy consumption and material use. The energy and N_2 gas consumption were multiplied for the specific emission factors corresponding to electricity (kg CO_2 eq/kWh) and N_2 gas (kg CO_2 eq/m^3) production (Ecoinvent, 2024; European Environment Agency, 2024).

3. Results

3.1. Samples characterization

The production scrap samples, both LFP and NMC cathodes, were provided by the National Research and Development Institute for Cryogenic and Isotopic Technologies (ICSI), Rm. Vâlcea, Romania. Their composition consists of 20 wt% current collectors and 80 wt% active material, with a ratio of cathodic powder, binder, and C65 of 92:5:3, reflecting the typical industrial cathode formulation used in commercial cells (Abdollahifar et al., 2023). The production scrap samples were characterized with XRD spectroscopy to identify the main crystalline phases and XRF and FAAS spectrometry to quantify the concentration of their elemental components.

The crystalline structure explored through XRD spectrometry included in LFP samples (Fig. 2A) the peaks of $LiFePO_4$ structure, and in NMC samples (Fig. 2B) the peaks of $LiNi_{0.8}Mn_{0.1}Co_{0.1}O$. The presence of PVDF as binder in both samples was confirmed by FTIR spectra (Supplementary materials Fig. S1), where the characteristic peaks of PVDF at 870 cm^{-1} and 1400 cm^{-1} had been identified (Bai et al., 2020b; Song et al., 2013).

The chemical composition of the samples (Table 1) revealed that Al impurities were detected: 822 ± 289 mg/kg in LFP and 12,542 ± 3706 mg/kg in NMC. The characterization of the LFP and NMC production scrap samples confirms the expected crystalline structures and binder presence, matching typical industrial compositions (Sobianowska-Turek et al., 2021; Xuan et al., 2019; Pagnanelli et al., 2024). The noticeable difference in aluminum impurity levels between the LFP and NMC samples may reflect differences in manufacturing processes or material degradation, which could affect recycling methods and efficiency for these cathode materials.

3.2. Recycling efficiency

The temperatures and atmosphere environments tested were specifically chosen to explore the effect of thermal treatment on both the PVDF binder and the active materials, with particular focus on their thermal stability and recovery potential, as summarized in Fig. 3. Thermal detachment was tested at 200 °C, just above the binder melting point; at 300 °C, a temperature between melting and full degradation; at 400 °C, which exceeds the binder degradation temperature; and at 600 °C, the standard temperature for complete binder degradation. For higher temperature (400 °C and 600 °C), tests were conducted in both air and inert atmospheres to examine the impact of iron oxidation. Particularly for LFP, thermal treatment above 400 °C must be conducted under an inert atmosphere to prevent oxidation of $LiFePO_4$ to $Li_3Fe_2(PO_4)_3$ and Fe_2O_3, with oxidation state changing from Fe (II) to Fe (III) (Lai et al., 2023), and making the material unsuitable for direct recycling in air.

The achieved recovery efficiencies (Fig. 4) clearly indicate that process temperature is a key parameter influencing the recovery of active materials. The highest recovery was achieved for both types of scrap cathodes after thermal treatment at 600 °C, confirming that thermal treatment at this temperature is highly effective in completely separating the active materials from the current collector (Bruno and Fiore, 2025; Ji et al., 2023). For LFP samples, the thermal treatment process is highly effective, with the recovery of both Li and Fe improving significantly as the temperature increases (Fig. 4A). At 400 °C, 98 ± 7 % of Fe and 99 ± 11 % of Li were recovered after treatment in air, while in an inert atmosphere, slightly lower values of 95 ± 6 % for Fe and 84 ± 13 % for Li were observed. These results suggest that applying N_2 gas

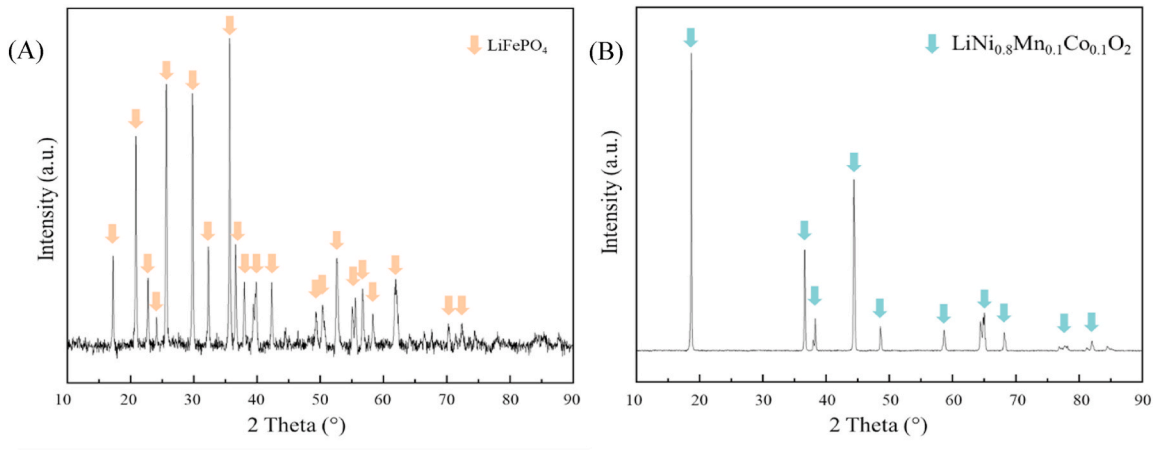

Fig. 2. XRD spectra of active materials form (A) LFP production scraps and (B) NMC production scraps.

Table 1
Elemental composition of the active material in the samples of LFP and NMC cathodes production scraps.

Sample	Li (%wt.)	Fe (%wt.)	Co (%wt.)	Mn (%wt.)	Ni (%wt.)
LFP	3.99 ± 0.34	29.89 ± 1.41	–	–	–
NMC811	6.19 ± 0.37	–	5.54 ± 0.56	1.77 ± 0.18	31.44 ± 8.44

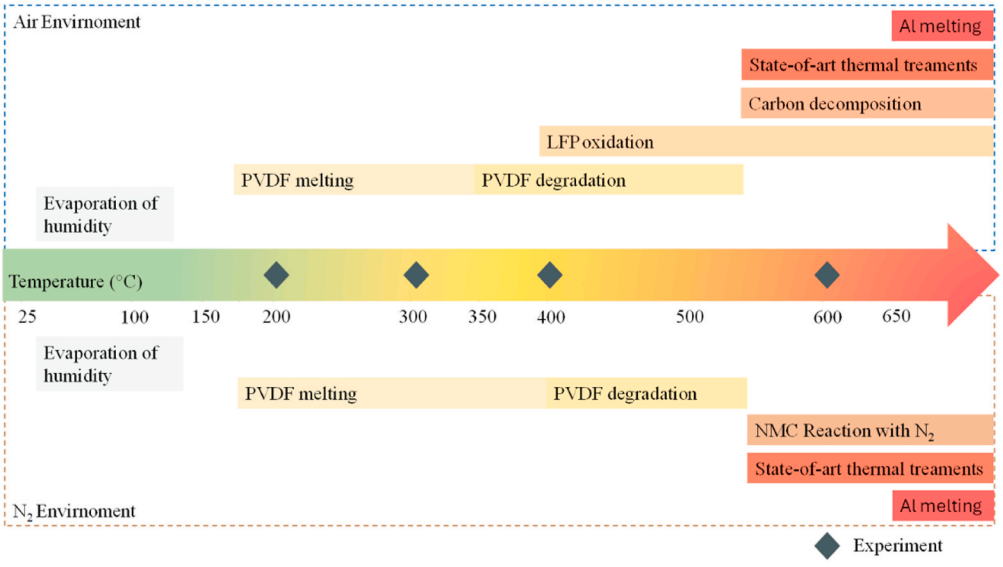

Fig. 3. Summary of the thermal behaviour of PVDF and LFP and NMC active materials in air and inert atmosphere at different temperatures.

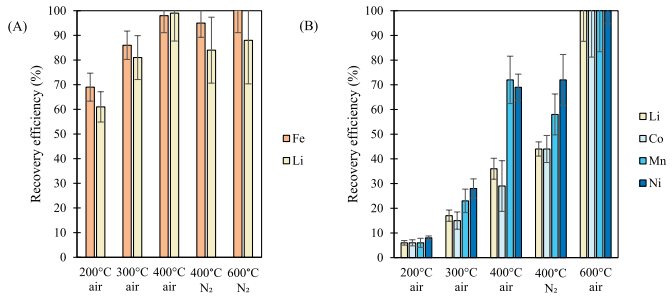

Fig. 4. Recovery efficiency after thermal treatment and ball milling applied to (A) LFP and (B) NMC production scraps.

flow may hinder the full release of the active material due to increased activation energy of thermal degradation of PVDF in inert atmosphere (Lombardo et al., 2021). NMC samples, instead, showed limited recovery efficiency at lower temperatures. At 200 °C, the recovery efficiency remains limited to 6 ± 1 % for Co, 6 ± 1 % for Li, 6 ± 2 % for Mn and 8 ± 1 % for Ni, indicating that this temperature is insufficient to decrease binder adhesion and to separate the active materials. As temperature increased to 400 °C, recovery efficiency improved significantly across all metals, especially Mn (72 ± 10 %) and Ni (69 ± 5 %).

Considering the effect of thermal treatment at different temperatures on the crystalline structure of the samples, the behaviour of LFP and NMC materials differed significantly at high process temperatures. The XRD spectra of LFP samples in N_2 environment revealed that the crystalline structure remained intact across all temperatures, showing no significant changes (Fig. 5A). However, when the thermal treatment was

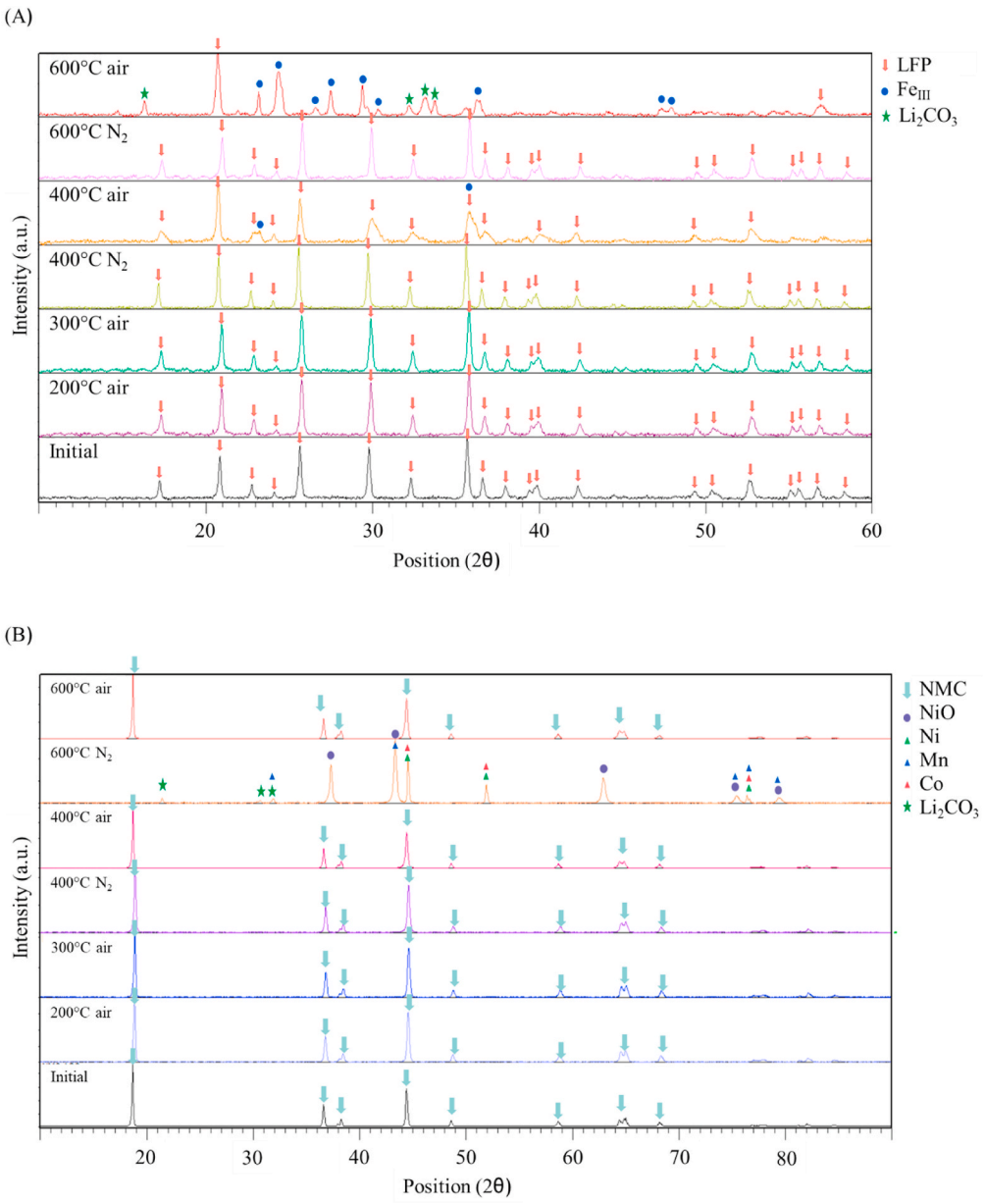

Fig. 5. Comparison of XRD spectra of (A) LFP and (B) NMC cathodes samples treated at different temperatures in air and inert atmosphere (N_2).

conducted in air at temperature above 400 °C, oxidation phenomena became evident, as indicated by the appearance of peaks corresponding to Fe_{III}, and changes in the intensity and shape of the main diffraction peaks (101), (111), (211), and (311) suggesting the formation of less ordered regions within the crystal lattice and partial structural degradation (Zhu et al., 2018). At 600 °C in air, the XRD patterns of LFP showed further deterioration, with most of the identified diffraction peaks corresponding to higher iron oxidation states, indicating that the original LFP structure has been destroyed. In contrast, NMC cathodes required an oxidizing atmosphere at high temperatures to maintain the integrity of the crystalline structure. The XRD spectra of NMC samples treated in an air environment exhibited minimal changes, with the main diffraction peaks remaining largely even after treatment at 600 °C. Thermal treatment under inert conditions resulted in structural degradation, which hinders material recovery (Lombardo et al., 2021). The XRD patterns revealed a complete degradation of the crystalline lattice, after treatment at 600 °C under inert conditions, accompanied by the formation of metallic components and metal oxides, which aligns with previous studies (Liu et al., 2018). Specifically, high temperature thermal treatments lead to redox reactions among the current collector, the active carbon and the NMC material during which the transition metals in the active material are reduced from higher to lower oxidation states (Yang et al., 2016).

Another critical factor influencing recovery efficiency is the purity grade of the recovered materials, which is affected by the presence of Al impurities from the current collectors (Supplementary materials Fig. S2). In LFP samples, Al impurity content remains consistently low (<1100 mg/kg) across the entire temperature range and under both process atmospheres. However, a clear trend emerged, with Al contamination decreasing at higher temperatures. The lowest Al concentration (55.93 ± 13.83 mg/kg) was observed in the LFP sample treated at 600 °C in N_2. In contrast, higher Al concentrations were detected at lower temperatures, particularly in the range between the PVDF melting point and its degradation temperature. Notably, Al content was higher after treatment at 300 °C (1074.33 ± 212.99 mg/kg) than at 200 °C (310.91 ± 26.33 mg/kg), suggesting that the melting of the binder strengthens the adhesion between the active material and the current collector, hindering effective separation. NMC samples, on the

other hand, exhibited significantly higher Al contamination at lower temperatures compared to LFP. At 200 °C Al content reached 47,897 ± 2517 mg/kg and it increased to 51,475 ± 83.65 mg/kg at 300 °C, which are however below the Al concentrations reported after solvent-based detachment processes (Ji et al., 2024). Similarly to the non-linear trend observed on LFP materials, this suggests that intermediate temperatures, while partially facilitating recovery, can exacerbate impurity levels due to incomplete binder decomposition (Hanisch et al., 2015), posing a significant challenge for direct recycling processes. Such contamination must be considered in practical process design, particularly when the target is high-purity material suitable for direct reuse in cathodes. Nevertheless, previous studies suggested that limited traces of Al may enhance the electrical conductivity of recycled cathode materials, potentially mitigating the impact of such contamination in certain applications (Hanisch et al., 2015; Lei et al., 2024; Zhang et al., 2020).

Therefore, optimal thermal treatment conditions for efficient material recovery from both LFP and NMC scrap cathodes are temperature-dependent and significantly influenced by the atmosphere. For LFP, treatment at 400 °C in air provided the highest recovery efficiency for both Li and Fe, achieving nearly complete recovery (98 ± 7 % for Fe and 99 ± 11 % for Li). However, to prevent Fe oxidation, it is essential to adopt an inert atmosphere and temperatures at least of 400 °C. For NMC, higher temperatures are critical for effective recovery, and treatment at 600 °C provided the highest recovery rates for all metals. However, an air atmosphere was essential at this temperature to avoid structural degradation and preserve the integrity of the material. In contrast, when an inert atmosphere was used at higher temperatures, the crystalline structure of NMC underwent degradation, which significantly hinders the recovery process. In summary, the recycling of LFP CAM presents a clear trade-off between recovery efficiency and material integrity. While 400 °C in air achieves the highest recovery, the XRD analysis confirms partial oxidation of Fe^{2+} to Fe^{3+}, compromising the crystal structure. Conversely, operating at a milder 200 °C in air yields slightly lower but still substantial recovery (~70 %) while fully preserving the LFP crystal structure.

3.3. Economic analysis

The total gas consumption was calculated by considering the sum of the time required to reach the process temperature with a heating rate set at +5 °C/min, the 30 min dwell time at the target temperature and the time required by the muffle to cool down to 200 °C before retrieving the samples, whit an approximate rate of −4 °C/min. Specifically, for treatments at 400 °C under an inert atmosphere, N_2 gas was supplied for 52.5 min, corresponding to a consumption of 105 L of N_2, while at 600 °C the N_2 supply duration increased to 142.5 min, corresponding to a request of 285 L. When scaled to the maximum treatment capacity of the muffle furnace at laboratory scale, the total N_2 gas consumption amounts to 2100 L to process 1 kg of samples at 400 °C and 5700 L at 600 °C. The energy demand was directly measured during experimental testing and normalized considering the maximum treatment capacity of the laboratory equipment, including the muffle furnace and the planetary ball mill.

The energy demand associated with the thermal treatment enlarged proportionally to the increase in process temperature, corresponding to 1.92 kWh to process 1 kg of scrap sample at 200 °C, 3.32 kWh at 300 °C, 5.42 kWh at 400 °C and 10.42 kWh at 600 °C. The choice of air or inert atmosphere during the thermal treatment did not significantly impact the overall energy demand. Additionally, the energy consumption for milling the samples was measured as1 kWh per kg of processed material. Factoring in the market prices for energy and N_2 gas (see section 2.3), the processing costs were calculated corresponding to the expenses required to treat 1 kg of LFP and NMC production scraps (Supplementary materials Table S1). Notably, while increasing the process temperature led to a higher energy demand, the use of inert conditions had much greater impact on overall processing costs. For instance, applying an inert N_2 atmosphere at 400 °C raised the processing cost from 1.00 €/kg (in air) to 2.18 €/kg, and at 600 °C from 1.62 €/kg (in air) to 4.82 €/kg. This indicates that the cost associated with N_2 gas supply is a more significant driver of total process cost than temperature increase alone. These material and energy requirements apply equally to LFP and NMC samples since they refer to the operational needs to process 1 kg of scrap material, reflecting the physical and thermal conditions necessary for processing but they do not consider the process efficiency. Therefore, the processing costs to treat 1 kg of samples was converted into the recovering costs to obtain 1 kg of active material, accounting recovery efficiency for LFP and NMC samples (Fig. 6).

The higher recovery efficiency obtained for LFP samples, led to lower recovery costs ranging between 0.71 €/kg LFP after treatment at 200 °C and 4.82 €/kg LFP after treatment at 600 °C in inert atmosphere. While the recovery efficiency is maximal at higher temperatures, XRD analysis shows that Fe oxidation occurs, making 200 °C in air economically and environmentally optimal while preserving material integrity. Whereas the recovery costs for NMC active material are affected by the lower recovery efficiency reported for these samples, and they fall between the range of 1.17 €/kg NMC after treatment at 400 °C and 4.22 €/kg NMC after treatment at 200 °C. For LFP, recovery efficiency remains relatively stable across different temperatures, meaning that increasing the process temperature mainly translate into higher processing costs without increasing significantly material recovery, thus raising the total recovery cost. It is important to distinguish between technical and techno-economic optima for NMC. While 600 °C in air achieves maximal recovery, this comes with higher energy use, gas consumption, and potential Al contamination. In contrast, 400 °C in air provides slightly lower recovery but minimizes energy demand, reduces costs, representing the most balanced and practical condition for industrial implementation. The minimum recovery cost corresponds to 0.71 €/kg LFP after treatment at 200 °C.Conversely, NMC materials exhibit an improvement in recovery efficiency with increasing process temperature, which partially offsets the rise in processing costs. A trade-off was identified for NMC at 400 °C in air, representing the optimal condition where recovery costs are minimized. Despite the inherently lower process efficiency of NMC compared to LFP, leading to higher overall recovery costs, it is important to note that for both types of scrap cathodes the recovery costs under all tested conditions remained significantly below the current market price of virgin cathode materials, which ranges between 12.57 €/kg and 14 €/kg for LFP or 25 €/kg and 29 €/kg for NMC (Gaines et al., 2021; Manjong et al., 2024; Orangi et al., 2024). These results highlight the economic viability of direct recycling for both LFP and NMC production scrap cathodes.

Based on the performed sensitivity analysis (see section 2.3) the most economically favourable condition for LFP was identified at 200 °C in air, for which the energy price could rise to 2.75 €/kWh while maintaining economic viability. While for NMC, the most cost-effective condition was 400 °C in air, allowing for an even higher admissible energy price of 3.89 €/kWh. Similarly, the highest fluctuation in market price of N_2 gas could reach 5.05 €/m^3 for LFP and 9.70 €/m^3 for NMC. These higher thresholds reflect the greater market value of NMC cathode materials (29 €/kg), enabling the process to absorb a higher energy on N_2 gas costs while remaining competitive with virgin material prices.

3.4. Environmental analysis

The energy and N_2 gas consumption required to process 1 kg of sample were converted into GHG emissions by applying emission factors corresponding to electricity (kg CO_2 eq/kWh) and N_2 gas (kg CO_2 eq/m^3) production, and then normalized by the recovery efficiency of each type of scrap material to calculate the CO_2 emissions required to recover 1 kg of active material (Supplementary Materials Table S2). This accounts for both the operational inputs (energy and N_2 gas) and the process performance, enabling a direct comparison of the environmental burden across different process conditions. Alongside the results of the

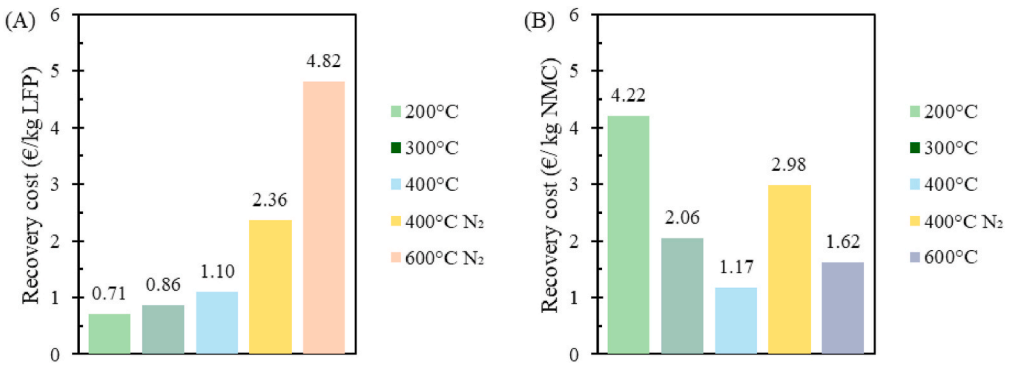

Fig. 6. Recovery costs (€/kg) for direct recycling of (A) LFP and (B) NMC cathodes production scraps.

economic analysis, the environmental impact was found to be influenced using an inert atmosphere; however, despite representing an additional impact item to the overall environmental burden, the increase in CO_2 emissions associated with N_2 use was comparatively less significant than its impact on processing costs. For instance, the processing emissions for treatments in air increased with temperature, from 0.75 kg CO_2/kg input at 200 °C, to 1.12 kg CO_2/kg at 300 °C, 1.66 kg CO_2/kg at 400 °C, and 2.69 kg CO_2/kg at 600 °C. When switching from air to inert conditions at 400 °C, emissions slightly increased from 1.66 kg CO_2/kg to 1.87 kg CO_2/kg. These results differ from the more than twofold increase in processing cost for the same change in atmosphere, indicating that while N_2 use should be carefully managed, its impact on GHG emissions was more moderate than its effect on the economic viability of the process.

To assess the environmental performance of each process route, the calculated processing emissions were normalized by the recovery efficiency, yielding the environmental impact per kg of recovered LFP or NMC active material (Fig. 7). The results reveal a clear distinction between LFP and NMC cathodes in terms of their environmental recovery costs. For LFP, the recovering GHG emissions increased steadily with temperature, from 1.18 kg CO_2 eq./kg LFP at 200 °C in air to 2.56 kg CO_2 eq./kg LFP at 600 °C in inert atmosphere. In contrast, NMC exhibited a different trend. At 200 °C in air, the environmental impact of recovery was substantially higher, reaching 6.98 kg CO_2 eq./kg NMC, primarily due to low recovery rates at this temperature. However, as the temperature increases, the emissions decreased significantly, dropping to 3.41 kg CO_2 eq./kg NMC at 300 °C, and further to 1.94 kg CO_2 eq./kg NMC at 400 °C in air. This strong decline is a direct result of improved material recovery efficiency, which offsets the additional energy demand. Notably, while 600 °C in air achieves the highest technical recovery (~100 %), 400 °C in air is the techno-environmentally optimal condition, balancing high recovery (~90 %), lower emissions, and manageable Al contamination. When an inert atmosphere was used at 400 °C, emissions increased slightly to 2.56 kg CO_2/kg and reached 2.69 kg CO_2/kg at 600 °C in N_2.

For direct comparison, literature data were analyzed as follows. When the results were reported as GHG emissions related to the recovery of the whole cells (kg CO_2 eq./kg of cells) or GHG emissions related to the cells' capacity (kg CO_2 eq./kWh), the values were converted in kg CO_2 eq./kg recovered material considering the average mass of active materials in LFP and NMC cells (Gaines et al., 2018) and the average specific capacity for LFP and NMC material (Reddy, 2011). A previous study on direct recycling of EoL LFP cathodes reported a very low environmental impact of 1.94 kg CO_2 eq./kg recovered LFP material through a low-temperature, aqueous relitihiation process that avoided thermal treatments (Xu et al., 2020). Similarly, in the case of NMC cathodes, direct recycling generated GHG emissions of 1.93 kg CO_2 eq./kg NMC, reflecting the benefit of avoiding energy-intensive thermal regeneration (Xu et al., 2021). In contrast, studies that employed high-temperature treatments and inert atmospheres reported considerably higher emissions, ranging from 3 to as much as 24.5 kg CO_2 eq./kg of recovered material (Chen et al., 2023). Even under more moderate operative conditions, emissions remained substantially higher than those reported in the present study, highlighting the environmental potential of material recovery through direct recycling of production scraps of LFP and NMC cathodes.

4. Discussion

4.1. Technical considerations

This study offers an effective, economically viable, and environmentally friendly solution for direct recycling production scraps from LFP and NMC cathodes, examining the characteristics of LFP and NMC production scraps and their behaviours under the tested conditions. At temperatures below 200 °C, the PVDF binder remained largely intact, preventing effective separation of the active materials from the current collectors as mentioned by literature (Wang et al., 2023). As the

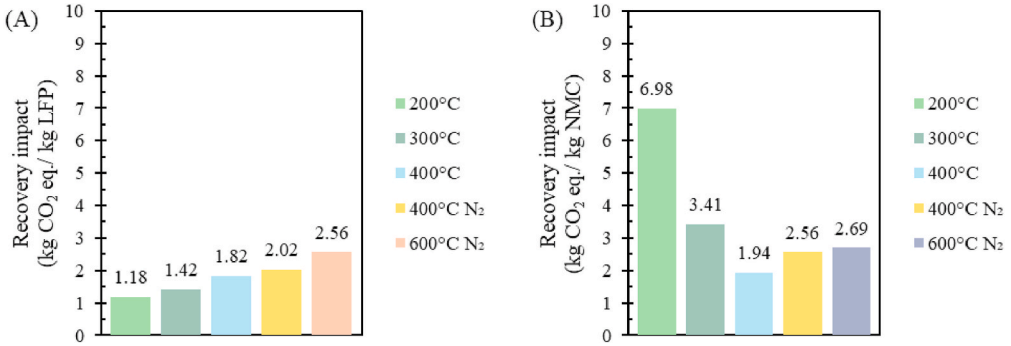

Fig. 7. Recovery impact (kg CO_2 eq/kg) for direct recycling of (A) LFP and (B) NMC cathodes production scraps.

temperature increased beyond 200 °C, the PVDF binder began to soften and melt, affecting the detachment of the active material from the collector. This phenomenon was more pronounced in NMC samples than in LFP, possibly due to differences in the binding mechanism of PVDF in NMC and LFP materials: PVDF had weaker binging force to the Al current collector in LFP than in NMC, and therefore the increase in the adhesion after the PVDF melting point has stronger effect on NMC (Zhong et al., 2021). When temperatures exceed 400 °C, PVDF degradation begins, potentially leading to the formation of undesirable by-products and a loss of electrochemical integrity in the recovered materials (Li et al., 2025). Moreover, higher temperatures, particularly those approaching 600 °C, led to partial structural degradation of the NMC active material, especially when processed in an inert atmosphere (Fan et al., 2024).

These preliminary observations have broader implications for industrial recyclers, particularly when processing mixed scrap streams containing both LFP and NMC. The differential behaviour of PVDF under thermal treatment suggests that a single processing temperature may not be optimal for all cathode types, requiring careful process design to maximize recovery efficiency while minimizing material degradation. Additionally, the technical trade-off between achieving effective separation and preserving material integrity must be considered in the context of alternative binder removal techniques. Methods such as solvent based processes or advanced thermal processes like pyrolysis can achieve binder removal at potentially lower temperatures or with different environmental footprints (Bai et al., 2020a; Wagner et al., 2023; Xu et al., 2020, 2021). Compared to these approaches, our low-temperature thermal/mechanical method is relatively simple, scalable using conventional industrial equipment, and avoids the need for high-pressure or chemically intensive processes, offering practical advantages in terms of operational simplicity and cost. The results of this study highlight the necessity of a technical trade-off required in thermal treatments for direct recycling, where careful temperature control is necessary to preserve both the integrity of the active materials and the efficiency of the recovery process.

Moreover, the identified conditions represent a local optimum within the limited parameter space explored, based only on discrete temperature levels and two atmospheric conditions. For more comprehensive optimization, future work should apply multi-variable statistical approaches, such as response surface methodology, to examine the combined effects of temperature, residence time, heating rate, and milling intensity. This would enable a more complete mapping of the process landscape and help determine a statistically validated optimum for material recovery, purity, and cost, as demonstrated in similar optimization studies (Tian et al., 2025).

4.2. Economic and environmental considerations

Both the economic and environmental analyses suggest that the most favourable conditions for direct recycling of LFP and NMC cathodes involve moderate temperatures (200 °C for LFP and 400 °C for NMC) in air. Operative conditions, such as processing temperature, atmosphere conditions, energy consumption, nitrogen gas use, and performance indicators, such as material recovery efficiency and purity of the recovered materials, provide a rationale behind the trade-offs between economic feasibility and environmental sustainability.

From an economic perspective, the key driver of processing costs is the use of inert atmospheres at higher temperatures (Roy et al., 2024). This suggests that while inert atmospheres may be necessary for preserving material quality, the added costs of nitrogen gas must be carefully managed. Thus, future optimization efforts should focus on minimizing the cost of nitrogen gas. This could involve technological advancements such as nitrogen recycling systems or the exploration of alternative inert gases that might be cheaper or more abundant (Harvey et al., 2022). Additionally, improving furnace efficiency and integrating renewable energy sources could reduce the overall energy demand, leading to lower processing costs while also mitigating the environmental impact. Nonetheless, these favourable economics assume an idealized, clean, and pre-sorted feedstock. In practical industrial settings, additional costs associated with sorting, separation, and handling of mixed or contaminated scrap streams could significantly influence overall process economics, as highlighted in recent reviews.

From an environmental perspective, the results indicate that the use of inert atmospheres has a less significant effect on GHG emissions, compared to energy emissions. Thus, one promising route for reducing environmental impacts is the integration of renewable energy sources. Solar, wind, and other low-carbon power sources could significantly reduce the CO_2 emissions associated with energy demand (Lin et al., 2023). Additionally, the use of less energy-intensive alternatives in the thermal treatment process could be explored. For instance, electric induction (Wagner et al., 2023) or microwave-assisted heating technologies, which are potentially more energy-efficient and can be powered by renewable energy, might be viable alternatives to traditional furnace-based heating.

4.3. Circular economy implications

The findings of this study suggest the strategic value of integrating direct recycling of LIB production scraps into manufacturing lines. Indeed, recovering active materials in situ reduces transportation costs and associated emissions (Gaines et al., 2021) but also overcomes scalability challenges by using conventional industrial equipment, without major capital investment. In the context of the European Union, which faces a shortage of primary raw materials and ongoing challenges in EoL LIBs management (Bruno and Fiore, 2024b; Gaines et al., 2023; Matos et al., 2022), this approach can substantially reinforce the security of the supply chain. Moreover, the proposed direct recycling process achieved economic viability, which is one of the major barriers to circular economy for LIBs. Given the low cost of virgin material, particularly for low-cobalt chemistries like LFP and NMC811, recycling must be cost-competitive (Atmaja et al., 2022; Lima et al., 2022). The demonstrated low-temperature approach not only reduces energy input but also preserves material integrity, offering a lower-cost alternative to other direct recycling methods.

However, further industrial-scale validation and the assessment of the electrochemical performance of the recovered materials would be required to accurately assess the validity of the proposed process. Phase integrity has been confirmed by XRD, and comparable studies in the literature indicate that similar low-temperature recovered powders can exhibit adequate electrochemical performance (Giles et al., 2024; Zhao et al., 2024). Future works should focus on full electrochemical validation, including initial specific capacity, coulombic efficiency, rate capability, and long-term cycling stability. Particularly the electrochemical evaluation of the recovered materials should entails re-slurring the materials into new electrodes, assembling them into coin cells, and assessing key performance metrics, including initial specific capacity, coulombic efficiency, rate capability, and long-term cycling stability. Such testing is especially important for the LFP sample treated at 400 °C in air, where XRD analysis suggested potential iron oxidation. Determining how this structural modification affects electrochemical performance is essential to fully validate the suitability of this processing condition.

Despite these challenges, the direct recycling process proposed in this study aligns with the EU's strategic goals to retain critical materials and limit the export of recyclable waste, positioning itself as a promising enabler of circularity within the battery value chain.

5. Conclusions

This study demonstrates the feasibility and benefits of direct recycling for LFP and NMC cathodes production scraps, operating standard industrial equipment and low energy demand. Technically, the process

proved effective in achieving high material recovery efficiencies, up to 98 ± 7 % for Fe and 99 ± 11 % for Li from LFP at 400 °C, and almost 100 % from NMC under the same conditions. The melting of PVDF at around 200 °C was sufficient to promote detachment of active materials without requiring full binder degradation, especially for LFP, enabling a milder, non-destructive recovery process.

The preliminary economic analysis reveals that the most cost-effective recovery conditions were achieved at 200 °C in air for LFP (0.71 €/kg) and 400 °C in air for NMC (1.17 €/kg). These values fall well below current market prices for virgin LFP and NMC materials, confirming the economic competitiveness of the proposed process. Sensitivity analysis indicated that energy prices could rise to 2.75 €/kWh for LFP and 3.89 €/kWh for NMC under the most favourable conditions while remaining cost-competitive, highlighting strong economic resilience. Whereas, from an environmental perspective, the direct recycling process also demonstrated promising performance. In air, GHG emissions per kg of recovered LFP increased from 1.18 kg CO_2 at 200 °C to 2.02 kg CO_2 at 400 °C, while NMC emissions dropped from 6.98 kg CO_2 at 200 °C to 1.94 kg CO_2 at 400 °C, reflecting improved recovery efficiency with temperature. When compared with results from previous studies, the present results are among the lowest reported for direct thermal recycling, especially because relitihiation is not necessary for direct recycling of production scraps.

In conclusion, this study confirmed the economic viability and environmental sustainability of direct recycling of LIBs cathodes production scraps. The simplicity and scalability of the direct recycling process makes it an excellent candidate for in-situ integration into battery manufacturing lines, particularly in regions like the EU, where resource circularity and material security are strategic priorities. Future work should focus on electrochemical performance validation of the recovered materials and techno-economic modeling at pilot or industrial scale to support broader deployment.

CRediT authorship contribution statement

Martina Bruno: Writing – original draft, Methodology, Investigation, Data curation, Conceptualization. **Lotta Liina Lassila:** Writing – original draft, Visualization, Investigation, Conceptualization. **Carlotta Francia:** Writing – review & editing, Validation, Supervision, Methodology, Conceptualization. **Annukka Santasalo-Aarnio:** Writing – review & editing, Supervision, Methodology, Conceptualization. **Silvia Fiore:** Writing – review & editing, Validation, Methodology, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.cesys.2025.100386.

Data availability

Data will be made available on request.

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