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Recent progress in pyrometallurgy for the recovery of spent lithium-ion batteries: A review of state-of-the-art developments

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Abstract

Pyrometallurgy is a well-known method for the efficient recovery of valuable metals from spent lithium-ion batteries (LIBs). This work provides an overview of the key aspects and recent advancements in pyrometallurgical processes for LIBs recycling. The newly developed pyrometallurgical processes have the potential to be energy-efficient, especially when utilizing microwave technologies. Despite encountering certain challenges and limitations, the prospects for recovering LIBs through pyrometallurgy appear promising, especially considering the anticipated rise in the number of spent LIBs for recycling.

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Pyrometallurgy, Metal recovery, Spent LIBs, Thermal treatment, Microwave technology.

Introduction

In our current era, marked by a pressing need for sustainable energy solutions, an increasing demand for portable electronic devices, and the electrification of vehicles, lithium-ion batteries (LIBs) have unquestionably become the leading energy storage technology [1,2]. Their widespread adoption is driven by their advantages, such as exceptional energy density, high specific power, absence of memory effect, low self-discharge rate, and a long lifespan. However, as their prevalence continues to grow, we are confronted with significant challenges, particularly in managing their disposal when they reach the end of their life cycle. Unsuitable disposal of spent LIBs not only presents safety hazards such as explosions but also contributes to environmental problems due to potential heavy metal and electrolyte pollution. This poses a threat to both the ecological environment and human health [3].

It is within this context that we recognize the paramount importance of addressing suitable recycling technologies, not only to protect the environment but also to ensure the responsible utilization and conservation of valuable resources, like Li, Co, and graphite, which are particularly concerning for LIBs, due to their geological availability, market constraints, and geopolitical factors. It's noteworthy that technology leaders in this field primarily concentrate their efforts on achieving high recovery rates for valuable elements such as Co, Ni, and Cu.

In this frame, a groundbreaking and transformative method has emerged as a promising approach in the realm of lithium-ion battery recovery: pyrometallurgy technology.

Pyrometallurgy is based on the use of high temperatures to recover valuable metals. At lower temperatures, the processes entail phase transitions and structural modifications, whereas at elevated temperatures, chemical reactions play a more prominent role [4].

Even if generally pyrometallurgical approaches are introduced to recover cathodic materials, more recent studies, microwave-based, also propose their use to treat anodic graphite [5]. Recent advancements in this field, offering the opportunity of shorter processing times and reduced energy consumption [6] hold the potential to produce fundamental advances in the recycling industry and significantly mitigate the environmental impact of retrieving spent batteries. In particular, the optimization of process time and energy is crucial in the context of recent changes in cathode chemistry, particularly the reduction of Co involvement.

The pyrometallurgy-based method is favored for commercial use due to its straightforward process, ease of scalability, and flexibility in handling various battery types [7]. In this review article, the pyrometallurgy

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processes for spent LIBs recycling are reviewed with great attention to the last results and improvements.

The pyrometallurgical recovery technologies

Pyrolysis and incineration

Pyrolysis is primarily used in the thermochemical decomposition of organic material.

In recent years, it has served as a key method for recovering both cathode and anode materials from LIBs. The most common operation in pyrolysis involves directly heating (with a temperature of more than 800 °C) the electrode materials in an inert atmosphere without any additional substances. The fundamental principle, when applied to a single electrode, revolves around heating to decompose, and vaporize the organic binder, consequently leading to the detachment of the cathodic and anodic materials from the corresponding metallic sheets.

Throughout the pyrolysis process, the organic materials (binders and electrolytes) within the electrode decompose, generating gases containing fluorine compounds [8], that can also be recovered by suitable systems [9]. In addition, pyrolysis allows to increase in the flotation efficiency of the electrode material due to the removal of organic materials [10].

Incineration, which is a thermal treatment with an oxidative atmosphere, can be also used as a preliminary step for LIBs recovery [11] to separate battery components and remove organic components. The optimal temperature of incineration is about 550 °C [12]. Both incineration and pyrolysis generate comparable quantities of fluorides. Nevertheless, incineration demonstrated a more significant environmental impact than pyrolysis, primarily attributable to its higher emission levels of CO and CO_2 [13]. On the contrary, if the objective is to achieve effective separation between the active material and the aluminum foil, the most cost-effective energy treatment is incineration. Incineration has demonstrated the ability to break down the organic binder in shorter durations and at lower temperatures compared to pyrolysis [13].

Smelting

In a conventional smelting procedure, high temperatures (1400–1700 °C) are applied to achieve the reduction of cathodic metals. Spent LIBs are considered such as naturally occurring ores and are heated with the addition of carbon, substances forming slag, and metal oxides simultaneously [7]. This process results in the formation of a Ni-Fe- Co-Cu alloy and a slag that contains Al, Mn, and Li. However, smelting suffers from notable drawbacks, including the loss of Li in the slag and the consumption of a substantial amount of energy [14]. Moreover, it is possible to increase the Li and Mn recovery, by substitution of the traditional smelting slag Al_2O_3—SiO_2-CaO with Al_2O_3—SiO_2-MnO or Al_2O_3—SiO_2-FeO [15]. Recently proposed studies showed that using an Al_2O_3-CaO slag [16], it is possible to recover Co, Ni, and Mn in the metal phase and simultaneously recover the Li in the flue dust as Li_2CO_3 and LiF.

Carbothermic reduction roasting

A more contemporary technique employed within the pyrometallurgical recovery process is carbothermic reduction roasting (CRR). The temperature, from 650 °C to 1000 °C, is used to promote reduction reactions of the metals from their oxides. This facilitates the conversion of materials containing strategic metals into chemically simpler forms, making them easier to separate. In carbothermic reduction, a carbon source is generally used to reduce the cathodic materials [17]. The fundamental process involves combining the electrode material (for example $LiCoO_2$ and $LiMnO_2$) with reducing agents (such as C and CO) or materials capable of generating reducing components (like biomass). These mixtures are subjected to a specific roasting temperature during the heating treatment. The process commences with the reactions of the cathodic materials and the additives. The decomposition of the additives yields reducing agents like CO. Simultaneously, the cathode material undergoes decomposition, resulting in the production of metal oxides (including Li_2O, CoO, MnO, NiO) and O_2 [4,18]. Ultimately, these oxides react with the reducing agents to generate individual metal components. CO_2 is generated by the reaction of C or CO with O_2. Li_2O can react with CO_2 and form Li_2CO_3 [18]. The strategic metals contained in the cathode are transformed into different phases that can be readily reclaimed using chemical dissolution or physical techniques.

In contrast to smelting, carbothermal reduction allows for a lower conversion temperature, reduced by 400–800 °C, as the thermodynamic equilibrium can be modified through the application of a vacuum or an inert gas atmosphere such as nitrogen [14]. One positive result of the CRR processes, compared with smelting, is that Li is converted into readily recyclable Li_2CO_3 or Li_2O rather than lost in slag.

Of the various reduction systems available (including urea, carbon-based, and melt-roasting systems), carbon reduction is the most widely diffused. In the anode and cathode mixed roasting setup, the primary reducing agents are carbon found in the anodic part of LIBs and the reducing gas generated as a consequence of the roasting process (CO). Then, currently, the mixed cathode and anode configuration is regarded as a valuable and financially viable approach for CRR. In addition to carbon thermal reduction, innovative CRR methods have been suggested, such as the exploration of biomass as a potential reducing agent for such applications.

Very recently, a carbothermic shock method was introduced to reduce a cathodic material with uniform temperature distribution, high heating and ultrafast reaction times [19]. In this case, the cathode is directly heated in a Joule-heating graphite boat, to be decomposed. Then, it is reduced by using the carbonized organic binder and native carbon as reducing agents.

The carbothermic reduction method was also proposed as an effective technique for regenerating spent anodic graphite, which can contain metals originating from batteries, such as Cu, Mn, Co, Li, and Al [20].

Salt-assisted roasting

This is an alternative roasting technique that transforms the diverse metals into water-soluble compounds. It can be classified based on the reagent employed as sulfation, chlorination, or nitration roasting. Salt-assisted roasting exhibits strong selectivity for Li and facilitates the separation of Li from transition metal oxides. Moreover, the reaction temperature is typically lower, from 200 °C to 1000 °C, compared to carbon roasting [14,21,22].

The fundamental process in sulfide roasting involves mixing the electrode material with sulfate-containing compounds (such as H_2SO_4, $NaHSO_4$, or $(NH_4)_2SO_4$) to initiate a sulfidation roasting procedure. In this process, cathodic metal ions like Co and Mn undergo initial reduction and subsequently combine with sulfate to produce metal sulfate. Additionally, Li ions can react to form Li_2SO_4.

Hence, the sulfurization roasting mechanism can be separated into two distinct steps. During the initial stage, the electrode material undergoes a reaction with sulfuric acid. Some metals (for example Mn, Ni, Co) experience reduction and sulfation. The following stage involves a solid-phase reaction with sulfuric acid at elevated temperatures. In the presence of carbon, the unstable $CoSO_4$ transforms into Co_3O_4, which then proceeds to undergo further reduction to CoO.

After roasting, the final materials that can be found are Co_3O_4, CoO, Li_2SO_4, and C [23].

The process of chlorination roasting closely mimics that of sulfidation roasting, involving the heating of electrode material while chlorinating agents are present (NH_4Cl, $CaCl_2$ and Cl_2) to obtain the conversion of cathodic metals [14]. The most commonly used chlorination roasting system is the one involving NH_4Cl. In this system, the roasting process is primarily governed by the solid-solid reaction (the interaction of $LiCoO_2$ with NH_4Cl) rather than the gas-solid reaction [21].

Most nitrates undergo rapid decomposition at lower temperatures (ranging from 125 to 250 °C), whereas lithium nitrate ($LiNO_3$) exhibits a considerably higher decomposition temperature of around 600 °C. The nitration recovery process involves converting the primary elements in LIBs scrap into their respective nitrates. These nitrates can be easily decomposed into insoluble oxides during roasting at appropriate temperatures ($<$300 °C), excluding lithium nitrates [24]. Subsequent water leaching of the roast enables the selective dissolution of lithium nitrate. The nitrogen oxide gases produced during roasting may be recycled to form nitric acid, which, in turn, may be reused in the initial nitration step of the proposed process.

Microwave-supported carbothermic reactions

The literature suggests that improving the sustainability of pyrometallurgical processes could involve substituting traditional energy consumption with renewable sources [25]. However, this recommendation may not consistently account for the fact that certain countries rely on coal for electricity generation [25].

In this frame, microwave (MW) radiation has been recently applied in various ways as new methodologies to recycle metals in spent LIBs. The researchers utilized anthracite as the reducing agent and carried out reduction roasting of electrode materials using MW energy [26]. The findings reveal that the cathode material exhibits excellent MW absorption capabilities within the range of 800−1200 °C. It is noteworthy that the dielectric constant of the cathode material experiences a sharp rise when the roasting temperature exceeds 600 °C, suggesting that MW energy can significantly enhance the efficiency of carbothermic reactions at temperatures exceeding 600 °C.

Simultaneously, it has been demonstrated that the CRR reaction is fully completed when the proportion of carbonaceous additives exceeds 18 % [26]. The findings indicate that the leaching rates of Mn, Ni, and Co, from products obtained through MW CRR exceed 97 %, with the leaching rate of Li reaching an impressive 99 % [27]. The incorporation of the MW heating technology enhances the pyrometallurgical approach and elevates the recycling opportunities for cathodic materials from LIBs.

However, despite that some research results demonstrate the potential of MW technology in the recycling of metals from LIBs, the activities in this field are still limited and new applications must be investigated. For example, MW technologies have been recently applied as new methodologies to recycle metals in lithium-ion batteries with a new approach, to promote carbothermal reduction reactions [26,28,29]. In particular, the use of a hybrid heating mechanism has shown promising results in the direct recovery of Li, which generally cannot be recovered by conventional pyrometallurgical processes [25].

Finally, MW treatment was also proposed to regenerate graphite, under an inert atmosphere [5].

The characteristics of pyrometallurgical technologies used for spent LIBs recovery are resumed in Figure 1.

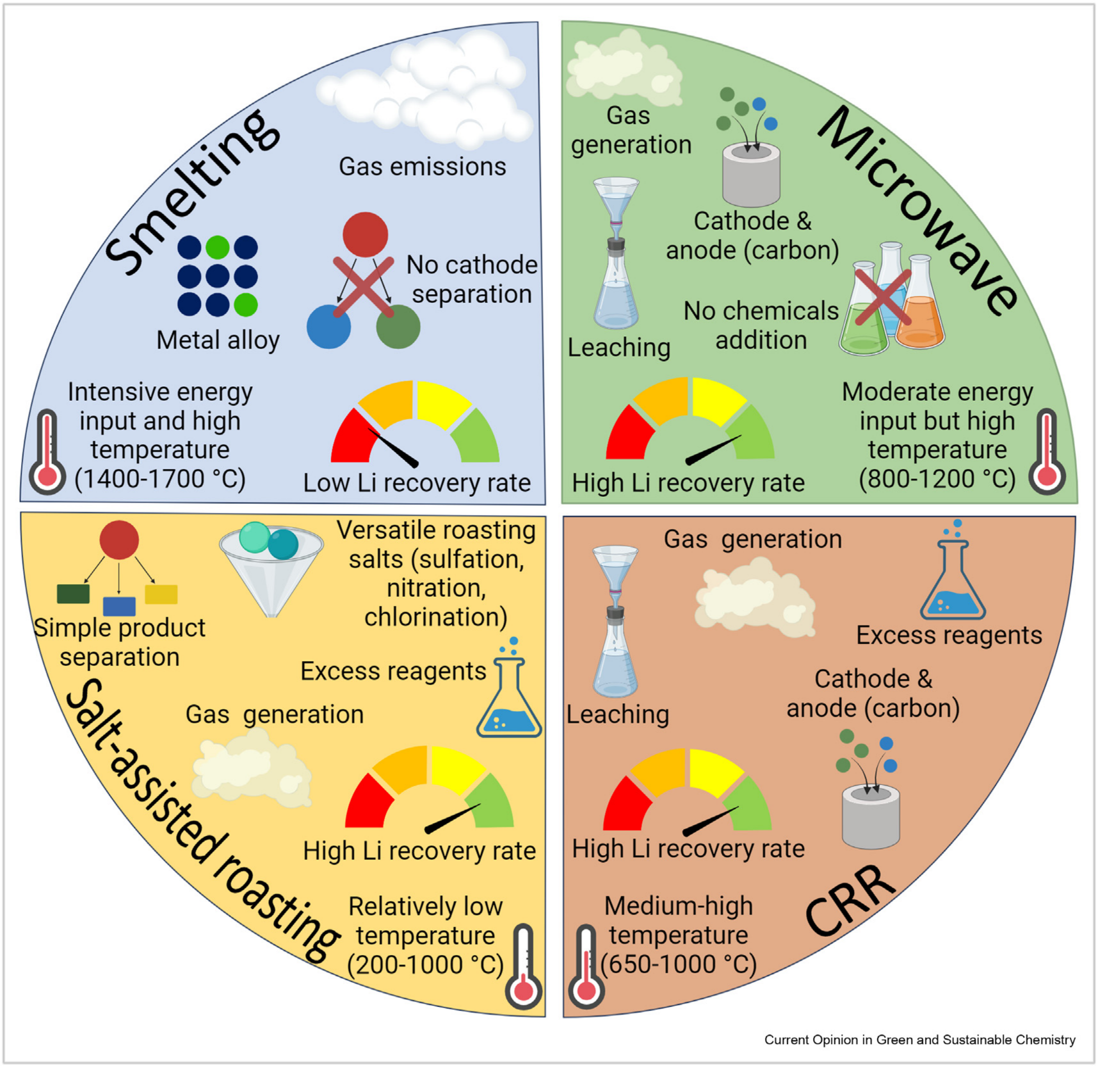
Advantages and disadvantages of the pyrometallurgical processes

Compared to other recycling methods, pyrometallurgy offers distinct advantages such as its ability to handle diverse battery chemistries, scalability, and energy efficiency when combined with modern technologies like MW heating.

The primary benefit of pyrometallurgical processes is that they eliminate the requirement for a preliminary raw material treatment step [30]. The pyrolysis recovery method efficiently accomplishes the separation of active materials from electrode sheets and resolves the challenge of reclaiming organic materials within electrode materials, achieving a safe retrieval of pyrolysis gases. Nevertheless, pyrolysis does not allow for the selective recovery of metals [31], and it needs significant dedicated and high-cost equipment. It also involves energy-wasting and the production of potentially explosive and toxic gases [32].

In particular, the application of CRR to the black mass can be realized without any thermal pre-treatment. Indeed, a great advantage of the CRR system is the existence of a carbon source in the black mass, due to the graphite presence, with no necessity for the addition of chemicals to support the reactions. However, there are still some disadvantages. Specifically, while the

Figure 1



Characteristics of different pyrometallurgical technologies used to treat spent LIBs for the recovery of strategic metals. CRR stands for carbothermic reduction roasting. The figure was created with BioRender.com.

Table 1

Temperature and atmosphere conditions, recovery of metals rate, advantages and disadvantages in technologies for metals recovery from spent LIBs.

	Temperature (°C)	Atmosphere	Recovery of metals	Advantages/disadvantages
Pyrolysis	300–900	Inert	No selective recovery	High-cost equipment, energy wasting, production of dangerous gases, long time treatment
Smelting	1400–1700	Air	Low Li recovery	Addition of metal alloys
CRR	650–1000	Air/Inert	Low Li recovery	No chemicals addition
Salt-assisted roasting	200–1000	Air/Inert	High metal recovery rate	Equipment corrosion, cost-effectiveness, low energy consumption
Microwave	800–1200	Air/Inert	High metal recovery rate	No chemicals addition, shorter time of treatment, still under development

carbothermic reactions can be operated at low temperatures, the overall metal recovery, particularly Li, may be less than that achieved with alternative methods [33].

A notable feature of sulfurization roasting is its high metal recovery rate, with most sulfide roasting systems achieving over 95 % recovery. However, with the exception of a few sulfurization roasting methods (such as the $(NH_4)_2SO_4$ roasting system, which operates at temperatures between 350 and 400 °C), most conventional sulfurization roasting processes necessitate temperatures ranging from 600 °C to 800 °C, indicating a generally high-temperature requirement. Additionally, the system of sulfurization roasting demands precise conditions for the reactions to prevent the formation of SO_x. Moreover, equipment corrosion represents a significant drawback associated with the sulfurization roasting system.

Chlorination roasting stands as one of the foremost methods for metal recovery from cathodic compounds and offers notable advantages for recycling spent LIBs. Firstly, most chlorination agents exhibit high chemical reactivity and swift chlorination rates. These attributes translate to elevated recovery efficiency and substantial metal extraction rates in the majority of chlorination roasting processes. Secondly, chlorinating agents are known for their wide variety and cost-effectiveness. Lastly, certain chlorination agents require roasting temperatures as low as 350 °C, contributing to reduced energy consumption.

Nevertheless, it is worth noting that some chlorinating agents, such as Cl_2, come with evident disadvantages due to their toxicity, necessitating appropriate measures to prevent leaks.

Finally, the potentialities and risks associated with MW processes have been not completely disclosed and they require more dedicated studies.

Table 1 reports the details of temperature, atmosphere conditions, recovery of metals rate, advantages, and disadvantages related to the different discussed methodologies.

Conclusion

Recent advancements in pyrometallurgy have shifted from smelting to roasting and from carbothermal reduction to salt-assisted roasting, to microwave technologies, with the aim of enhancing recycling efficiency and reducing energy consumption. Then, pyrometallurgy has been reconsidered because it stands as a promising solution for the recycling of spent LIBs, contributing to the development of a circular economy and boosting clean mobility. Continued research and development efforts in pyrometallurgy, coupled with increased awareness of e-waste's environmental impact, underscore the potential for further improvements in recycling efficiency and environmental benefits.

Author contributions

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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.

Data availability

Data will be made available on request.

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* * of outstanding interest

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