Review

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Toward Joule heating recycling of spent lithium-ion batteries: A rising direct regeneration method

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ABSTRACT

Lithium-ion batteries (LIBs) are critical for the rapid growth of electric vehicles (EVs), but their inherent lifespan leads to numerous retirements and resource challenges. The efficacy of conventional recycling techniques is increasingly compromised by their high energy consumption and secondary pollution, rendering them less responsive to greener and more sustainable requirement of rapid development. Thus, the direct recycling process emerged and was considered as a more expedient and convenient method of recycling compared to the conventional recycling modes that are currently in study. However, due to the reliance on the indispensable sintering process, direct recycling still faces considerable challenges, motivating researchers to explore faster, greener, and more cost-effective strategies for LIBs recycling. Inspiringly, Joule heating recycling (JHR), an emerging technique, offers rapid, efficient impurity removal and material regeneration with minimal environmental impact, addressing limitations of existing methods. This method reduces the time for direct recycling of spent LIBs by a factor of at least three orders of magnitude and exhibits significant potential for future industrial production. Unfortunately, due to the lack of systematic organization and reporting, this next generation approach to direct recycling of spent LIBs has not yet gained much interest. To facilitate a more profound comprehension of rising flash recycling strategy, in this study, JHR is distinguished into two distinctive implementation pathways (including flash Joule heating and carbon thermal shock), designed to accommodate varying pretreatment stages and diverse spent LIBs materials. Subsequently, the advantages of the recently developed JHR of spent LIBs in terms of material performance, environmental friendliness, and economic viability are discussed in detail. Ultimately, with the goal of achieving more attractive society effects, the future direction of JHR of spent LIBs and its potential for practical application are proposed and envisaged.

Keywords: Joule heating; Spent lithium-ion batteries; Flash recycling; Regeneration; Upcycling

Biography of authors

Haoxuan Yu is currently pursuing his MS degree at Nanchang Hangkong University. His research interests include battery recycling and reutilization and energy materials development.

Liang Chen received his Ph.D. degree (in 2017) in School of Chemistry and Chemical Engineering, Hunan University. He is currently a professor in School of Chemistry and Chemical Engineering, Hunan Institute of Science and Technology. His research interests mostly focus on recycling of lithium-ion battery and designing of energy storage materials and electrocatalysts. Until now, Prof. Chen, as an author/co-author has published over 80 papers in international SCI journals and owned 14 authorized national patents.

Liming Yang received his Ph.D. degree (in 2017) in School of Chemistry and Chemical Engineering, Hunan University. He is currently a distinguished professor in School of Life Sciences, Key Laboratory of Jiangxi Province for Functional Biology and Pollution Control in Red Soil Regions, Jinggangshan University. He has long been involved in research into clean recycling technology, theory and industrialization of end-of-life battery metals. Prof. Yang has published 62 SCI papers in international renowned journals (H-index=41, total citations=5340, 12 ESI Highly Cited Papers, 3 ESI Hot Papers, 1 Paper of the Year).

Xubiao Luo received his Ph.D. degree (in 2006) in School of Chemistry and Chemical Engineering, Hunan University. He has been committed to research on the principles, technologies and engineering applications of wastewater treatment and resource utilization for a considerable time. Prof. Luo has overseen the National Outstanding Youth, National Ten Thousand Talents Program, national key research and development project, and other projects. He has independently and cooperatively published more than 300 international SCI papers (including 25 ESI Highly Cited Papers) which have been cited more than 23,000 times with h-index of 80.

1. Introduction

Eliminating dependency on non-renewable energy and developing clean energy as an alternative is a promising approach to achieving the goals of "carbon peak and carbon neutrality" [1]. Lithium-ion batteries (LIBs), as representatives of advanced energy, are commercialized products designed to meet the stable operation requirements of electric vehicles (EVs). Driven by national policies, the global number of EVs is expected to grow to an astounding 125 million units by 2030 [2]. Clearly, the global demand for LIBs in the EV market is immense. However, with an average service life of 5–10 years, the limited operational lifespan of LIBs will lead to a large-scale tide of decommissioning [3,4]. Spent LIBs contain a significant amount of valuable metals, including Li, Ni, Co, Mn, etc. [5]. In the event of improper disposal, the presence of transition metals (such as Co, Mn, etc.) and organic pollutants in the environment has the potential to induce degradation in soil, water, and air, thereby posing a threat to human health [6-8]. Moreover, it is acknowledged that South America possesses more than 66% of the whole world's lithium reserves, with the remaining sources distributed across various regions, including North America, Australia, and Africa. This distribution poses significant challenges to mining and transportation logistics [9]. Given the extreme scarcity and uneven distribution of natural resources, coupled with regional policy restrictions, the potential supply pressure on these elements forces the recycling of spent LIBs thoroughly [10-14]. Currently, the main recycling methods include hydrometallurgy, pyrometallurgy, and direct recycling [15-17] (Fig. 1a). Among them, traditional recycling methods either suffer from high consumption of acids and alkalis or high energy consumption, which may no longer meet the stringent environmental governance standards of modern society [18-20] (Fig. 1b, c). Thus, the proper treatment of spent LIBs, based on safety and efficiency while considering both economic and environmental benefits, is a critical response to national policies and international regulations. Direct recycling, as an emerging method, stands out due to its short process, minimal secondary pollution, and high economic benefits [21-23].

From the perspective of direct recycling mechanisms, the focus for the cathode typically lies in element compensation and structural recovery, while for the anode, the removal of surface impurities and compensation for minor lattice defects are particularly important [24-29]. Accordingly, the regeneration processes for different active materials primarily include lithium supplementation via melting, solid sintering, inert gas pyrolysis, and non-thermal graphitization [30]. The treatment process can generally be divided into three simple steps: from failure process, to pretreatment, and then direct regeneration [31] (Fig. 1d). Although direct recycling has already seen significant improvements in convenience compared to traditional hydro-/pyro-recycling methods, there seems to be an unsatisfactory gap. Based on existing technologies, is it possible to find a faster, greener, and more cost-effective recycling strategy? Considering the transient heating characteristics of Joule heating and focusing on the physical and chemical property differences of various battery waste materials, Joule heating recycling (JHR) can quickly generate heat to remove impurities selectively in a very short time [32]. Simultaneously, the energy generated by the instantaneous current is converted into thermal energy, radiating onto the spent LIBs materials indirectly, which also benefits the regeneration process due to the rapid high temperature process [33]. It is anticipated that the rise of JHR could potentially push direct recycling for LIBs into a faster and greener new stage. However, despite the simplicity of these processes and the excellent performance of the regenerated materials, the application of Joule heat in LIBs recycling lacks systematic organization and positive reporting and has not yet sparked comparable interest (Fig. 1e).

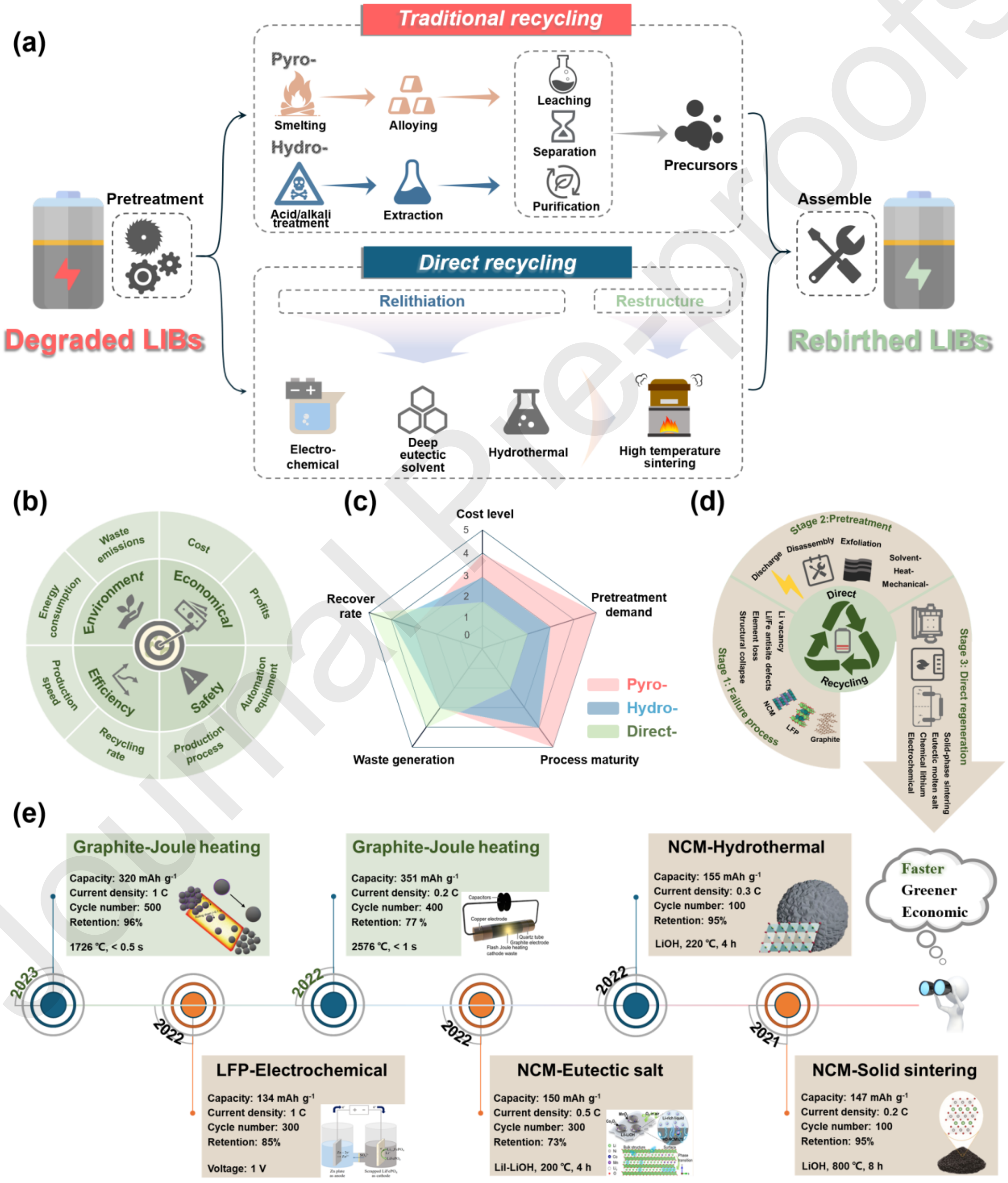

Fig. 1. (a) The three main recycling methods of spent LIBs. (b) Dimensions to focus on in recycling. Reproduced with permission from ref. [19]. (c) Advantages and disadvantages of the three recycling methods. Reproduced with permission from ref.

[20]. (d) Three step strategy of direct recycling. Reproduced with permission from ref. [31]. (e) Emergency and development history of JHR of spent LIBs.

Up to date, many battery recycling works have been largely reported. Xie et al. [34] presented a comprehensive review of the prediction of the whole-life health status of batteries, addressing key aspects such as data preprocessing, aging characteristics, and algorithms. In addressing this subject, certain teams have concentrated on closed-loop recycling strategies, conducting comprehensive analyses of sustainability studies on the entire life cycle of LIBs [30,35]. Among the strategies, direct regeneration has been mentioned extensively as both a frontier area and a pivotal issue. Consequently, this subject has given rise to a significant research surge, focusing on the concepts of repair and reutilization [36,37]. In addition, surveys have also been conducted on the topic of non-closed-loop recycling of LIBs [38,39]. And the objective is to facilitate the recycling of materials to meet the supply required for applications such as energy storage devices, sensors, catalytic reactions, etc. [40]. Gao et al. [5] directed their attention to the preferentially selective lithium extraction section, in which they analyzed five energy-driven lithium migration principles and underscored the initial breakthroughs and emerging advances in electrochemical lithium extraction of spent LIBs cathodes. The direct recycling section of spent LIBs, however, is conspicuously lacking an overarching assessment and promotion of the recently developed Joule heating recycling technology, which has come to be recognized for its high efficiency and short-duration requirements [41,42].

Herein, we first focus on the indispensable pretreatment phase, elaborating on the pretreatment processes that align with current mainstream recycling methods. We then classify the starting points for JHR, providing a detailed analysis of the two unique recycling paths (including flash Joule heating and carbon thermal shock) inherited from existing pretreatment methods. To meet the trends of universality and scalability, we systematically organize and evaluate JHR from the perspectives of material performance, environmental issues, and economic benefits, combining theoretical foundations with practical processes. Furthermore, given the growing demand for fast and economic recycling of spent LIBs, we also propose suggestions on how Joule heat strategies can make breakthroughs in the field of battery recycling.

2. Actualization of Joule heating recycling technique

In Joule heating technology, an electric current passes through an electrically conductive powder or film, the heat is generated due to the existence of resistance as internally. Therefore, contact Joule heat measure needs to have suitable conductivity for sufficient heat conversion to maintain the efficiency of the process ($P = i^2 \times R \times t$, where P is the power, i is the current flowing through the resister, R represents conductor resistance, and t represents the time of electrification). In addition to the electrical properties, it is essential to consider the thermal properties (thermal conductivity, specific heat capacity, etc.) and the mechanical properties, as these determine whether the equipment can achieve uniform in-situ heating and fine temperature regulation. Especially for contact heat measure of Joule heat, if properly designed, the influence of spatial variations on the heating result due to incorrect positioning can be minimized. Certainly, the heat generated from the Joule heat effect can also be transferred in a contactless measure. The unique carbon thermal effect of carbon-based material, due to its low specific heat capacity and high emissivity ($\varepsilon_{\text{carbon}}$ is 0.8–0.9 for visual wavelengths), even enables rapid heating and cooling with a sensitivity of 100,000 K s^{-1} within certain temperature ranges.

It is prominent to note that both contact and contactless measure necessitate the preliminary treatment of spent LIBs. To elucidate the application of joule heating recycling technology, the conventional pretreatment process is mentioned first, and two distinct pathways are delineated, contingent on the disparate material loading requirements. The focus is directed towards the emergent rapid recycling technologies that have been derived and optimized from conventional recycling methodologies.

2.1. Pretreatment

Before recycling spent LIBs, a pretreatment phase must be carried out. For spent power batteries from EVs, which are typically composed of numerous battery modules, these battery packs may maintain relatively high voltage and capacity as usual. Even minor collisions or local short circuits can easily cause the battery to become uncontrollable, potentially leading to fires or explosions [43-45]. To prevent risks such as short circuits and explosions, and to facilitate the subsequent recovery of active materials, the spent LIBs materials must, in general, undergo three treatment steps: passivation, disassembly, and separation, unless otherwise specified.

2.1.1. Passivation

To reduce risks, aqueous solutions are commonly used for passivating the LIBs [19] (Fig. 2a). During short circuiting of anode and cathode, residual energy is almost completely released through the process of electrolysis of water. Saturated NaCl solutions are considered to be cheap and mild conductive solutions, widely used in both laboratory and large-scale discharging [46]. High concentrations of NaCl can reduce discharge time, but excessive salt solutions may corrode the battery casing and internal electrodes. This may result in leakage of waste liquid and the release of toxic gases [47,48]. Techniques such as gel adhesion and semiconductor assistance have introduced innovations to both liquid-phase and solid-phase passivation methods, improving the protection of the electrode plates and accelerating the current, thus optimizing the discharge process for better material quality and efficiency [49,50]. Furthermore, strategies like freeze-curing strategies are being developed to promote electrolyte failure and achieve passivation of LIBs. Therefore, it is crucial to balance electrode corrosion, acid consumption, and the thermal energy generated during discharge to lay the groundwork for the subsequent recycling process.

2.1.2. Disassembly

After discharging the battery, the first step in facilitating separation is to remove the individual modules from the battery pack [19] (Fig. 2b). Considering the components of the battery, including the casing, cathode plates, anode plates, separator, and electrolyte, manual disassembly is regarded as the preferred method for achieving precise separation. However, high labor costs and low production rates greatly hinder the scalability of battery disassembly. Consequently, mechanical disassembly methods have emerged as a development opportunity and are increasingly favored by industrialized LIBs recycling companies. Nevertheless, the existing pyrolysis-mechanical disassembly-crushing-sorting processes are still insufficient to meet the demand for high-purity recovery of individual components. The material flow separation technology for spent LIBs urgently requires reform and breakthroughs.

2.1.3. Separation

Following the disassembly step, the current collectors coated with active materials, which hold high economic value for both the anode and cathode, are typically a focal point in the recycling process [19] (Fig. 2c). The active materials must be separated from the current collectors to efficiently collect the active black powder. However, during the battery manufacturing process, the addition of binders such as polyvinylidene fluoride (PVDF), styrene-butadiene rubber (SBR), and carboxymethyl cellulose (CMC) was necessary to prevent the shedding of active materials, which undoubtedly presents a challenge for the separation and recovery of the active substances. Solid-phase thermal treatments at high temperatures (300–600 °C for 1–3 h) can effectively pyrolyze organic binders, facilitating separation [51]. In liquid-phase processes, solvents can be used to dissolve current collectors, organic binders and active materials, utilizing alkaline solutions for dissolution, deep eutectic solvents (DES), and polar solvents (e.g., NMP) can be used for soaking to achieve separation [52-54].

2.2. Joule heating recycling strategies

2.2.1. Flash Joule heating

Flash Joule heating (FJH) refers to flash Joule heat process that involves transient high temperature treatment of the anode/cathode powders after fine separation by separation step (Fig. 2d). Three prominent features need to be considered during its application. (1) Capacitor effect: the instantaneous movement of charge triggers direct electrification of the material, enabling precise adjustment of ultra-high heating power (>3000 K) on a millisecond scale. (2) Conductivity: semiconductor carbon-based materials assist conductivity, and the local temperature differences caused by resistance variations facilitate directional impurity removal. (3) Contact heat: the current induced by the capacitor is transmitted instantaneously through the instrumental wires, resulting in the occurrence of thermal effects directly in the powder material. For the cathode, although the residual PVDF after separation step is locally decomposed at high temperatures, the extremely short duration of FJH may not fully restore the material from the surface to the interior. The anode generally uses water-soluble binders, which leave only trace amounts after separation. In this case, FJH mainly focuses on restoring the graphite layered structure and improving the electrochemical performance at high temperatures. Therefore, the effectiveness of FJH recycling depends on the properties of the spent LIBs powder, requiring no additional devices or reagents, thus achieving short-duration, low-energy regeneration of active materials.

2.2.2. Carbon thermal shock

Without deep separation, carbon thermal shock (CTS) refers to the rapid recycling that directly processes the electrodes separated during disassembly step. Three prominent features need to be considered during its application. (1) Power drive: this method relies on continuous energy input to heating the current collectors. (2) Contactless heat: the external carbon cloth, carbon fiber, and other thermal conductive carriers are heated through thermal conduction or radiation. (3) Application field: this technology is generally employed in the industrial synthesis of alloys and carbon materials, with the high yields and process stability, while simultaneously destroying the binder and high temperature restructuring of the active materials. However, due to the specific properties of the cathode current collector (Al foil), excessively high temperatures may reduce its toughness and cause cracking, making material separation more complex. It may be possible to find a balance between PVDF failure and preserving the integrity of Al foil by altering the atmosphere and adjusting the temperature control, thus achieving efficient separation of cathode active materials. For the anode, Cu foil can maintain a certain structural strength at high temperatures, and with the thermal decomposition of the water-based binder, the graphite anode can successfully separate from the current collector, compensating for some structural defects. Therefore, this CTS recycling seems more suitable for processing graphite anode and possesses the universal characteristics needed for large-scale applications.

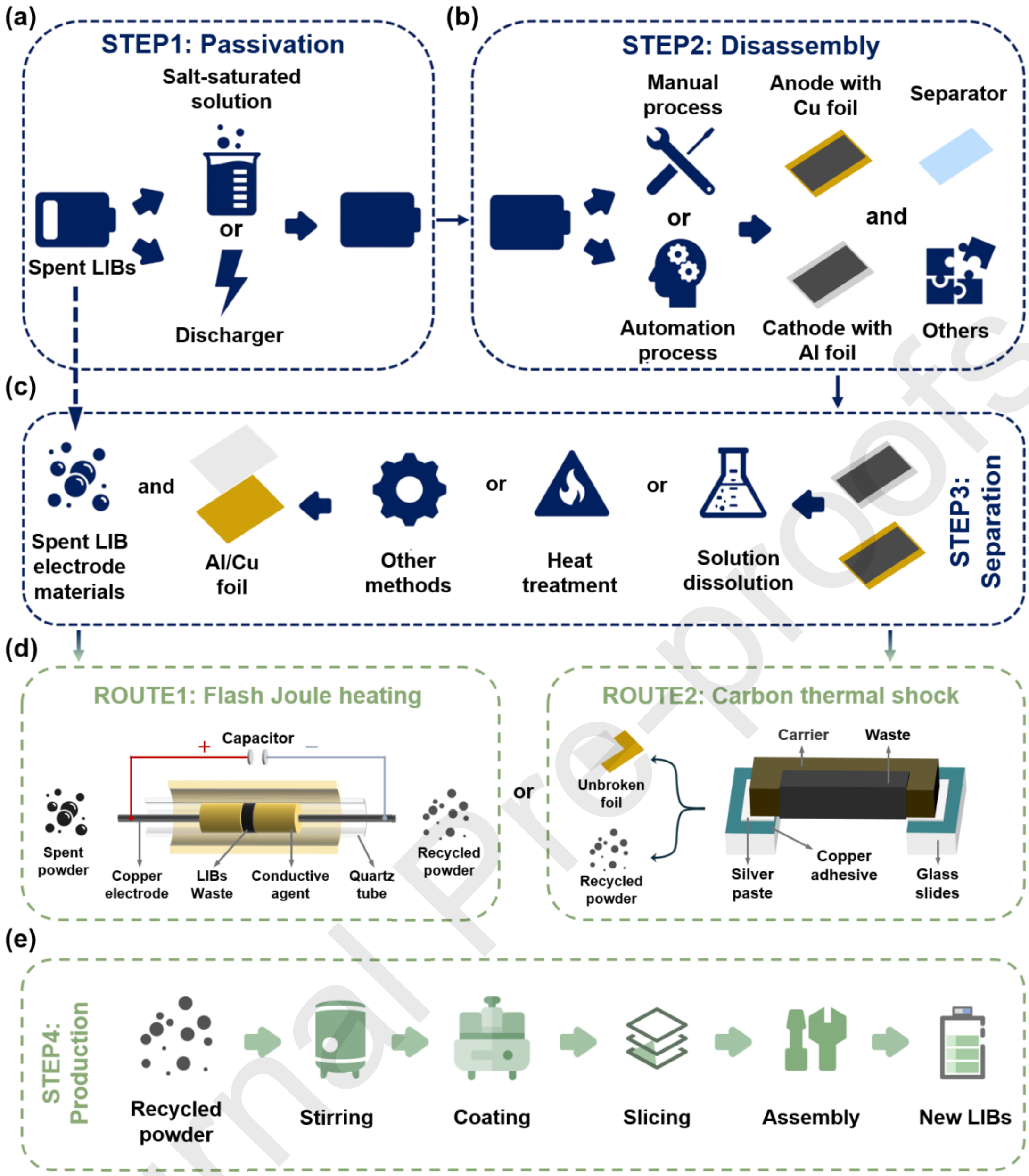

Fig. 2. The pretreatment of LIBs recycling including (a) passivation, (b) disassembly, and (c) separation. Reproduced with permission from ref. [19]. (d) Flash Joule heating and carbon thermal shock recycling routes of JHR. (e) The scheme of LIBs production after the JHR process.

Finally, the active materials recovered from the above two recycling routes will be used in an entirely new attitude to serve the production of LIBs through the crafts mainly including slurry preparation, coating, slicing and assembly (Fig. 2e).

3. Forays of Joule heating for LIBs recycling

3.1. Rapid recycling of graphite anode

3.1.1. Regeneration

As the most used anode material in LIBs, graphite is often overlooked in recycling processes due to its relatively low value. However, the JHR method meets the temperature requirements for both impurity decomposition and phase transitions, offering a promising new route for the recycling of spent graphite anodes.

Chen et al. [55] developed an ultra-fast flash method for treating graphite anode waste (Fig. 3a). The FJH system rapidly heats up to 2800 K in 3 ms, with the Joule heat effect inducing the decomposition of the resistive solid electrolyte interface (SEI) layer while simultaneously promoting the formation of an amorphous carbon shell (Fig. 3b and c). Finite element simulations demonstrate uniform high temperature distribution during the flash process, and the rapid heat dissipation of the system also helps avoid graphite anode expansion and defect formation. After flash recycling, residual metal oxide nanoparticles (Li, Co, etc.) can be efficiently recovered using 0.1 M HCl. The regenerated graphite (RG) anode retains its original three-dimensional layered graphite core structure, and the assembled anode material achieves a specific capacity of 351.0 mAh g^{-1} at 0.2 C with initial coulombic efficiency (ICE) of ~80% and maintains 77.3% of its capacity after 400 cycles at 0.5 C (Fig. 3d and e). Given the harsh inert atmosphere conditions, one may question whether Joule heating still effectively regenerates graphite in normal air conditions. Dong et al. [56] improved the Joule heating process by developing a constant pressure air atmosphere rapid temperature control device (reaching 3000 K within 0.1 s). The binder, SEI components, and lithium embedded between graphite layers rapidly evaporate at high temperatures, and after cooling, the RG exhibits an ordered layered structure (I_d/I_g = 0.103) (Fig. 3f). The RG anode material demonstrates excellent rate performance (350 mAh g^{-1} at 1 C) and cycle performance (99% of capacity retention after 500 cycles at 1 C).

Thus, regeneration of spent graphite can be achieved in both inert and air atmospheres, and the electrochemical performance of the regenerated material approaches commercial standards. Moving into the second stage, the question arises: how can the processing efficiency be increased and its applicability broadened to meet future potential large-scale use? Li et al. [57] developed a slanted carbon heating recycling process, utilizing gravity to replace horizontal transmission (Fig. 3g). When the degraded graphite rolls down the heater, continuous high temperatures of 2000 K within 0.1 s can remove surface impurities and improve graphitization (expanded 002 interlayer spacing). The RG anode material shows a reversible capacity of 320 mAh g^{-1} at 1 C, with 96.0% of capacity retention after 500 cycles. Despite this slanted carbon heating process attempting to leverage gravity to reduce energy consumption and offering a feasible recycling solution, obtaining refresh powders still necessitates the process of separation step, which complicates the overall process. In response, Zhang et al. [58] developed a continuously rolled-over heating recycling process (Fig. 3h). The integrated continuous carbon thermal impact device can directly process graphite waste (anode electrodes), rapidly peeling graphite while simultaneously repairing the material (Fig. 3i and j). The regenerated graphite anode material shows a capacity of 340 mAh g^{-1} at 0.5 C, with over 98% of capacity retention after an additional 180 cycles, rivaling commercial graphite (CG) (Fig. 3k).

3.1.2. Upcycling

JHR, as a unique rapid heating method, typically offers the ability to modify material properties. For graphite anodes, techniques such as element doping, coating layer modification, and interface engineering are widely considered beneficial for enhancing material performance. Cheng et al. [59] focused on addressing defects in spent graphite anodes and proposed a targeted repair-upgrading strategy. Due to differences in binding energy, Sn is more likely to combine with graphite defects rather than other sites (Fig. 3l). The team employed a roll-to-roll scalable technology with horizontal transmission for element supplementation and directional regeneration, using Sn ($SnCl_2$ as a precursor) dispersed through FJH (reaching 1873 K within 50 ms) to nucleate in graphite defects (Fig. 3m). The nanoscale dispersion of Sn helps increase lithium intercalation speed and capacity. The upgraded RG anode material shows an ICE of 84.3% and a specific capacity of 458.0 mAh g^{-1} after 100 cycles at 0.2 A g^{-1}. Ji et al. [60], considering the formation of the SEI layer during electrochemical cycling and the accumulation of residual active lithium, developed a rapid heating strategy (reaching 1900 K within 150 ms) that instantly transforms the loose original SEI layer into a dense inorganic layer that coats the graphite surface (Fig. 3n–p). The reconstructed SEI and encapsulated active lithium endow the regenerated graphite with excellent ICE, which significantly improves the ICE (98.8% vs. 83.2%) and energy density (309.4 vs. 281.4 mAh g^{-1}) when compared to CG in full cell configuration with LFP.

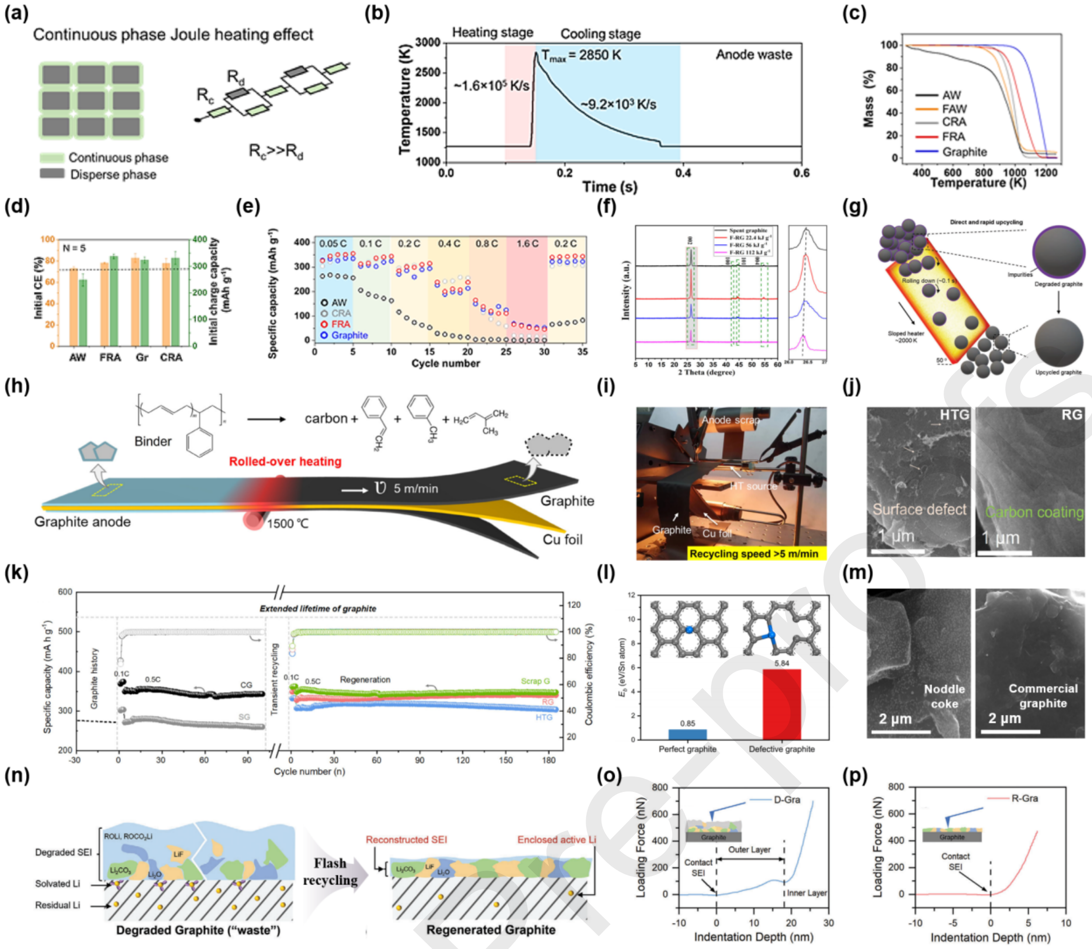

Fig. 3. (a) Resistance-dependent Joule heating in multiple phase systems. (b) Procedures of FJH recycling of graphite anode and (c) the thermogravimetric analysis (TGA) curves of different graphite anodes. (d) Results of ICE and charge performance and (e) the rate capacity of different samples. Reproduced with permission from ref. [55]. (f) X-ray diffraction (XRD) patterns of flash recycled graphite and spent graphite. Reproduced with permission from ref. [56]. (g) Schematic of sloped carbon heater for direct rapid recycling graphite anode. Reproduced with permission from ref. [57]. (h) The sketch and (i) the digital photo of rolled-over heating strategy. (j) Scanning electron microscope (SEM) images of high temperature and RG anode and (k) cycling performance of different samples. Reproduced with permission from ref. [58]. (l) Binding energy of single Sn atom on fresh and defective graphite and (m) SEM images of the CG and needle coke after nucleation. Reproduced with permission from ref. [59]. (n) Flow diagram exhibiting the transformation of degraded graphite to RG and the indentation curves of the (o) degraded graphite and the (p) RG. Reproduced with permission from ref. [60].

3.2. Rapid recycling of cathode waste

3.2.1. Regeneration

Regenerating cathode materials is particularly challenging, with factors such as atmosphere, temperature, and time requiring precise control. The JHR, a rapid high temperature solid-state calcination process, may not fully restore the crystal structure within the reduced time. However, its potential for direct regeneration could still offer valuable insights. Chen et al. [61] developed a solvent-free, anhydrous FJH recycling method combined with magnetic separation to recover fresh cathodes from waste materials, achieving solid-state reduction (Fig. 4a). To assess the versatility, several different cathode/anode materials were compared in parallel under FJH treatment (Fig. 4b and c). The cathodes exhibited a complete core structure and layered characteristics, demonstrating substantial potential for reconstruction into new cathodes. Based on this, Yin et al. [62] designed a roll-to-roll ultra-fast high temperature manufacturing process, which can rapidly relithiate and repair the crystal structure of degraded LCO powder within 8 s (Fig. 4d). The X-ray diffraction (XRD) results indicate that the regenerated LCO after flash recycling process does not result in the formation of spinel Co_3O_4 compared with that of spent LCO, exhibiting layered oxide structure with $R\overline{3}m$ space group (Fig. 4e). The regenerated LCO cathode material achieves an initial discharge capacity of 133.0 mAh g^{-1} at 0.1 C, with excellent cycle performance for over 300 cycles.

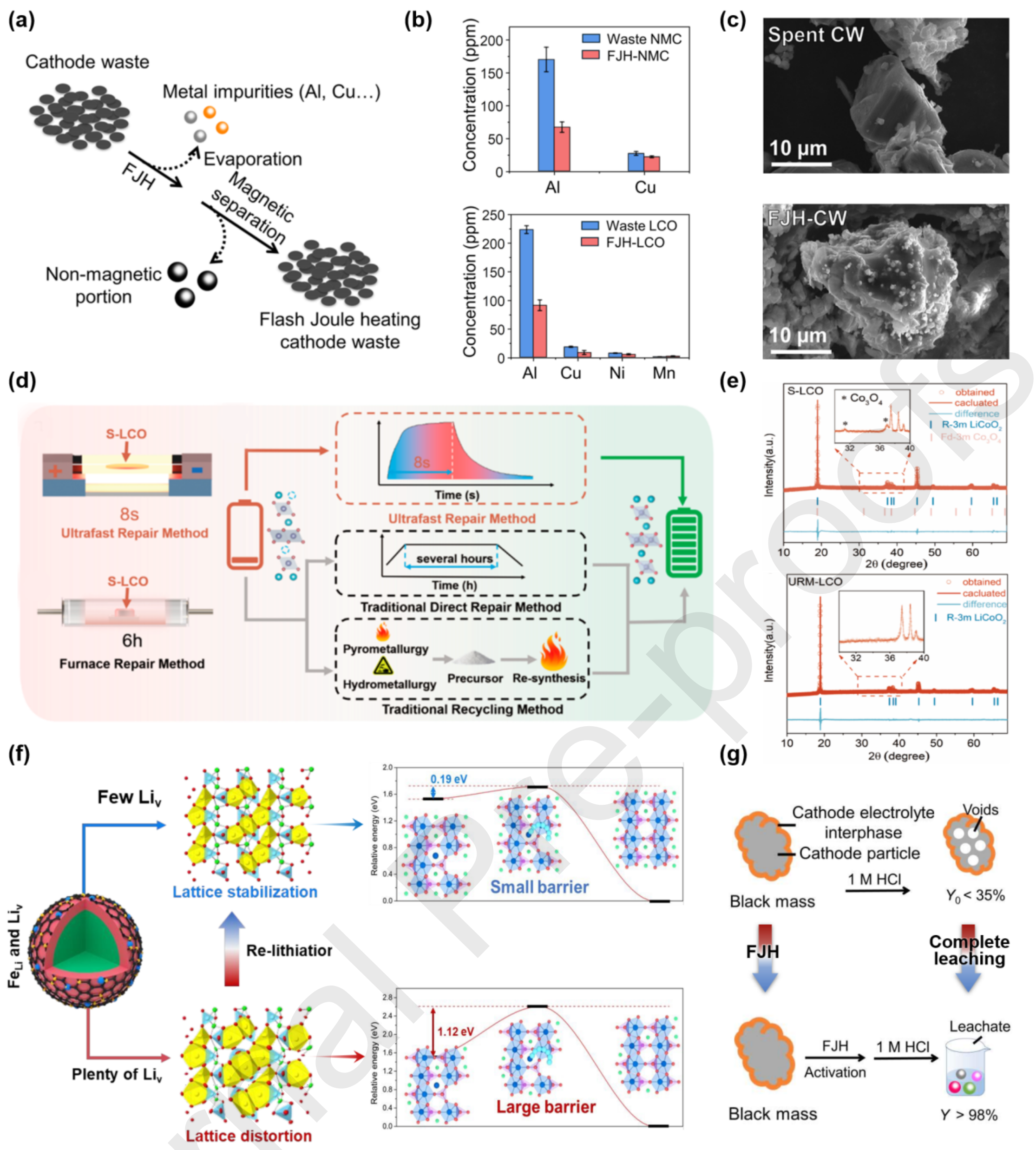

Fig. 4. (a) Schematic of flash recycling of cathode waste (CW). (b) The concentration of impurity metals in waste LCO and FJH-LCO. (c) SEM images of spent CW and FJH-CW. Reproduced with permission from ref. [61]. (d) Flow chart of ultrafast recycling process and furnace regeneration method. (e) XRD Rietveld refinement results for S-LCO and regenerated LCO. Reproduced with permission from ref. [62]. (f) Schematic representation of the energy curve and FeLi site. Reproduced with permission from ref. [63]. (g) The acid leaching results of direct leaching and FJH activation leaching process. Reproduced with permission from ref. [64].

3.2.2. *Upcycling*

Considering residual impurities such as PVDF, amorphous carbon, and graphite in the cathode waste, can the Joule heating regeneration process direct some organic components into other forms that may play a role in the regenerated cathode material?

Guo et al. [63] employed CTS to impact spent LFP powder, manipulating the atomic structure to achieve rapid, non-diffusive lattice jumps, repairing Li-Fe antisite defects without damaging the material integrity (Fig. 4f). Meanwhile, the PVDF remaining on the material surface was directed through the CTS process to form a fluorine-doped carbon coating that adhered to the surface of the LFP particles. The residual inorganic lithium salts combined with the carbon coating to form a composite cathode electrolyte interphase (CEI) film, significantly improving the material’s electronic conductivity and lithium-ion transport rate. The restored LFP cathode material exhibited an ICE of 97.0% and demonstrated a capacity retention of 97.6% after 300 cycles at 1 C.

3.2.3. Element extraction

Therefore, the JHR method offers certain benefits in impurity removal and repair for spent LIBs cathodes. However, the reduction in energy consumption and time also limits its ability to achieve optimal direct regeneration, resulting in the need for subsequent high temperature sintering treatments. Considering the JHR process’s ability to nanosizing and homogenize metal particles, combining it with traditional hydrometallurgical methods may lead to new discoveries. Chen et al. [64] applied FJH treatment to spent black powder (both cathode and anode), heating it to 2100 K in a few seconds, which facilitated the decomposition of the SEI and reduction of metal compound oxidation states. After rapid thermal treatment, the organic impurities that previously hindered leaching on the surface of the black powder were broken down, creating unobstructed extraction paths, resulting in a dramatic 1000-fold improvement in leaching kinetics (Fig. 4g).

3.3. Faster and more suitable recycling for manufacturing

As illustrated in Table 1, a macroscopic comparison of the Joule heat application and previous direct regeneration methods is provided in the field of crafts and product performance [21,23,28,55,57,58,60–63,65–68]. It is evident that both of Joule heating recycling routes facilitate the rapid regeneration of materials, resulting in substantial time and cost savings when compared to previous regeneration means such as leaching, sintering, and annealing.

In terms of graphite anodes, the specific capacity of recycled graphite is generally lower than that of direct regenerated products. Nevertheless, it has been demonstrated to be applicable in numerous scenarios pertaining to energy storage. Furthermore, due to the lattice reconstruction and impurity evolution driven by transient high temperatures, recycled graphite can offer unique properties due to the residual nano-groups frequently, such as high CE, well lithium diffusion rate, and significant electrochemical activity.

In the context of layered cathodes, it has been observed that the attainment of complete recovery of the layered structure is rendered challenging by the presence of instantaneous high temperatures. In comparison with conventional direct regeneration, the initial discharge capacity of Joule heating recovered LCO is lower and difficult to exceed 150 mAh g^{-1}, suggesting that there is considerable scope for further research on rapid performance recovery.

In the case of olivine-structured cathodes, which possess a more robust lattice structure than layered cathodes, the concentration of elemental deficiencies is observed to be on active lithium. Concurrently, the spatial skeletons of PO_4 tetrahedra and FeO_6 octahedra remain intact. Thus, Joule heat technology recycles LFP primarily for the purpose of rapid relithiation. It is encouraging to reveal that Joule heat technology has the capacity to restore the electrochemical performance of LFP to a level that is comparable to that of fresh commercial LFP materials.

Table 1

Summary of direct recycling of spent LIBs and corresponding performance comparison.

No.	Type	Technology	Time	Performance	Ref.
1	Graphite	FJH	<1 s	Durability: 0.2 C Cycle: 120 Capacity: 351.0 mAh g^{-1}	[55]
2	Graphite	CTS	~0.1 s	Durability: 0.2 C Cycle: 500 Capacity: 350.0 mAh g^{-1}	[57]
3	Graphite	CTS	<1 s	Durability: 0.5 C Cycle: 180 Capacity: 340.0 mAh g^{-1}	[58]
4	Graphite	FJH	~0.15 s	Durability: 0.2 C Cycle: 100 Capacity: 321.5 mAh g^{-1}	[60]
5	Graphite	Leaching Annealing	>9 h	Durability: 1.0 C Cycle: 450 Capacity: 355.0 mAh g^{-1}	[23]
6	Graphite	Leaching	>4 h	Durability: 0.1 C	[65]

		Sintering Polymerization		Cycle: 100 Capacity: > 350.0 mAh g^{-1}	
7	LCO	FJH Annealing	~13 h	Capacity: 142.0 mAh g^{-1} Cycle number: 100 Durability: 0.2 C Capacity retention: 96.9%	[61]
8	LCO	CTS	8 s	Capacity: 123.9 mAh g^{-1} Cycle number: 300 Durability: 0.2 C Capacity retention: 80.7%	[62]
9	LCO	Leaching Sintering Annealing	>24 h	Capacity: 142.1 mAh g^{-1} Cycle number: 100 Durability: 0.5 C Capacity retention: 97.4%	[66]
10	LCO	Sintering Annealing	>20 h	Capacity: 160.2 mAh g^{-1} Cycle number: 100 Durability: 1.0 C Capacity retention: 91.2%	[67]
11	LFP	CTS	1 s	Capacity: 152.8 mAh g^{-1} Cycle number: 300 Durability: 1.0 C Capacity retention: 97.6%	[63]
12	LFP	CTS	20 s	Capacity: 104.0 mAh g^{-1} Cycle number: 400 Durability: 2.0 C Capacity retention: >99.0%	[28]
13	LFP	Annealing	>15 h	Capacity: 157.0 mAh g^{-1}	[21]

				Cycle number: 400 Durability: 5.0 C Capacity retention: 88.0%	
14	LFP	Hydrothermal	>2 h	Capacity: 156.0 mAh g^{-1} Cycle number: 100 Durability: 2.0 C Capacity retention: >90.0%	[68]

4. The feasibility of Joule heating recycling

In the last few years, there has been a growing recognition of the urgent need to develop large-scale recycling markets for power batteries, due to constraints imposed by both resource and environmental factors. The feasibility of this development hinges upon a dual assessment of economic sustainability and process maturity. The JHR, as an emerging recycling method, has gained attention due to its short time and low energy consumption, which suggests considerable potential for further development and application. The present study aims to assess the feasibility of JHR for processing spent LIBs by evaluating the recovery efficiency of value components (mainly including cathode and anode materials) based on typical research cases and different recycling pathways.

4.1. Advantages of anode recycling

4.1.1. With carbon thermal shock

As shown in Fig. 5(a), the CTS recycling process, represented by roll-to-roll transient recycling, is compared with traditional recycling methods. Pyrometallurgy typically yields metal oxides and alloys and fails to recover graphite anode, while hydrometallurgy is effective in obtaining pure graphite but requires substantial reagents and numerous energy inputs. In contrast, CTS recycling can instantly heat LIBs anodes (<1 s), simultaneously recovering both graphite and intact copper foil. This process is highly convenient and demonstrates extraordinary time and energy efficiency. According to the EverBatt model, the energy and material inputs for all three recycling methods are mainly composed of energy and materials inputs, with CTS recycling requiring no additional reagents and a treatment cost as low as 2.48 MJ kg^{-1} cell, which is only 2% of that required for hydro-recycling (Fig. 5b). Additionally, waste emissions exhibit a similar trend, with the emission index for the three recycling methods shown in Fig. 5(c). It is evident that CTS recycling has the lowest emissions, reducing them by 97% compared to hydro-recycling. Therefore, this recycling method eliminates the complex pretreatment to obtain powder materials and uses instantaneous thermal energy to separate and repair the graphite in spent LIBs anodes. The process is notable for its low energy consumption, time cost, and carbon emissions, suggesting significant potential for future large-scale applications.

4.1.2. With flash Joule heating

Similarly, the FJH recycling process, represented by direct rapid upcycling, eliminates the need for acid use and long calcination times, resulting in a much shorter process compared to traditional recycling methods (Fig. 5d). After appropriate separation and cleaning, spent graphite is directly loaded onto the Joule heating equipment, and no auxiliary reagents are required. The instantaneous current induced by charge movement across the capacitor terminals is converted into thermal energy, and a millisecond (~150 ms) high temperature treatment facilitates beneficial evolution of the graphite from the surface to the core. The primary economic benefits and costs are composed of factors such as materials, energy inputs, process efficiency, and disassembly. Direct rapid upcycling demonstrates exceptionally low costs and high revenues, primarily due to its simplified process and the high value of the resulting products (Fig. 5e and f). Notably, the energy input for direct rapid upcycling is mainly derived from precise disassembly and separation, with the process standing out due to its minimal water usage and low gas emissions. Compared to hydro-recycling, this rapid regeneration method exhibits significant benefits in both economic (reducing costs by approximately 68%) and environmental (reducing emissions by approximately 79%) metrics (Fig. 5g–i).

4.2. Advantages of cathode recycling

Undoubtedly, the recycling of cathode materials from spent LIBs has been a focal point of research due to their high economic value. FJH, as an initial attempt to pave new pathways for LIBs recycling, has shown that treating only graphite anodes is clearly insufficient. Could flash recycling of cathodes also offer similar economic benefits or trends? In this study, we use the flash recycling-separation-resynthesis process as a case for FJH recycling, where graphite anodes serve as original additives to increase conductivity, with subsequent resynthesis based on solid-state sintering (Fig. 5j). Through a multi-dimensional parallel comparison of energy consumption, material use, and carbon emissions, flash recycling demonstrates reductions of 83%, 62%, 72%, and 58% in water use, energy consumption, gas emissions, and cost, respectively, compared to hydro-recycling (Fig. 5k–p). Furthermore, when compared to current direct recycling processes, FJH recycling shows improvements across all dimensions. As interest in low-cobalt content cathode materials grow, these ferromagnetic wastes may benefit from FJH recycling, yielding higher recovery rates and economic returns.

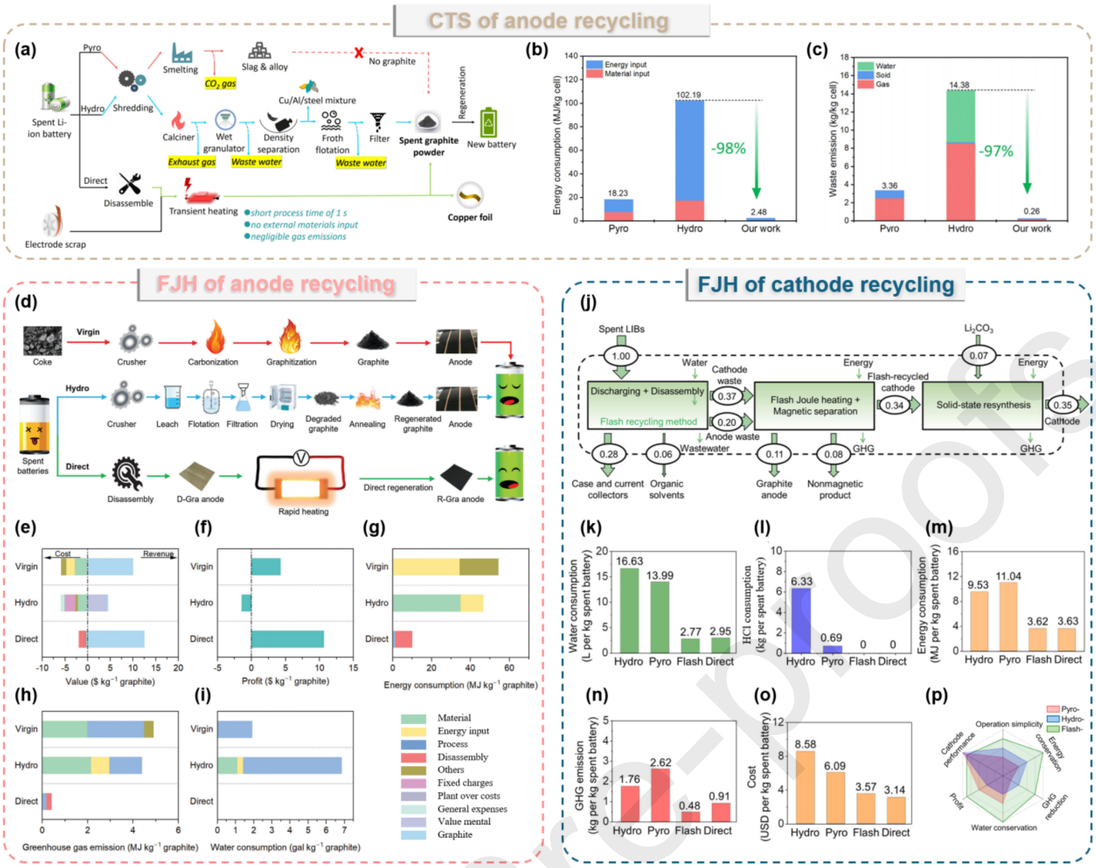

Fig. 5. Economic and environmental analysis of different recycling methods. CTS recycling for anode. (a) Comparison of different recycling processes. (b) Energy consumption and materials input and (c) waste emissions. Reproduced with permission from ref. [58]. FJH recycling for anode. (d) Flow chart of three main recycling methods. (e) Cost and revenue. (f) Profit. (g) Energy input. (h) Greenhouse gas (GHG) emission. (i) Water consumption. Reproduced with permission from ref. [60]. FJH recycling for cathode recycling. (j) Flow diagram of flash recycling method. (k) Acid consumption. (l) Water consumption. (m) Energy consumption. (n) GHG emission and (o) the total cost. (p) Comparison among different recycling processes. Reproduced with permission from ref. [61].

4.3. Potential limitations

4.3.1. Material selection

For FJH, the material resistance impacts the heat generation and energy utilization of system. Lower resistance results in increased current consumption, potentially placing significant strain on system components. Conversely, high resistance affects the smooth current transportation, hindering the effective excitation of the joule heat effect. Consequently, the selection of lower current and higher resistance is an effective measure for space-saving and management enhancement. In the context of black mass obtained from spent batteries, the incorporation of additives such as conductive carbon and graphite prior to FJH is imperative to augment the ultimate conversion utilization of energy. Moreover, the employment of metal components is widespread due to their exceptional thermal conductivity, thereby ensuring that the fundamental requirement of uniform heating of the system is met. Examples of such metals include nickel, chromium, iron, and alloys. However, chromium, tungsten, and molybdenum typically exhibit low efficiency and are difficult to scale up due to high cost. Therefore, FJH recovery has been heavily limited in fundamental investigations to explore the dynamic process of rapid repair of spent LIBs material. In the case of CTS, the material may not be loaded into a quartz tube, and the current does not pass directly through the material. It is more likely that the transient high temperature treatment of the material is achieved by using a metal or carbon material with high thermal conductivity as a medium. Notably, the loading of carbon-based materials (parasitic heat losses, higher radiant power density) has led to a new breakthrough for CTS.

4.3.2. Energy control

In addition to the heating means and material selection, there are fundamental discrepancies in the energy supply between FJH and CTS, which will result in divergent developmental trajectories. The FJH model stimulates the Joule heat effect through the transfer of electrons initiated by the immediate release of capacitance. Conversely, the CTS model relies on a direct current or alternating current power supply, which is more readily available and necessitates less equipment.

$$(\lambda,T) = \gamma\varepsilon_{\mathrm{gray}}\frac{2hc^2}{\lambda^5}\frac{1}{e^{hc/\lambda K_{\mathrm{B}}T}-1} \tag{1}$$

Where K_{B} is the Boltzmann constant, h is the Planck constant, c presents the light velocity, λ presents the wavelength, $\varepsilon_{\mathrm{gray}}$ denotes the stable emissivity, and γ denotes the fitted constant. In pursuit of continuous improvement and with the objective of enhancing the performance and sustainability of the process, several strategies have been proposed. These include the adoption of a periodic switching model, an intensive heating strategy and a roll-to-roll approach, as well as the preparation of support-free, homogenous and large-scale nanomaterials by combining atomized precursor with CTS. These efficient and energy-saving methods have undoubtedly had a significant potential impact on the development of rapid recycling of spent LIBs.

5. Summary and outlook

This work presents the Joule heating recycling process as a recent advancement in the direct regeneration of spent LIBs. It offers a multidimensional and systematic comparison with traditional hydrometallurgy and pyrometallurgy. This study analyzes the inheritance of Joule heating recycling from traditional pretreatment to subsequent flash recycling process. Furthermore, it elaborates on the scope and effectiveness of this emerging method for different battery wastes. It is evident that Joule heating recycling provides a range of advantages over direct regeneration and hydrometallurgy, including enhanced efficiency, economic viability, and environmental benefits. However, it is important to acknowledge the challenges associated with the authenticity of the restored material and its acceptance by the industry, the incompatibility of the recycled material with multiple applications, and the need to balance processing time and energy consumption (Fig. 6). These factors, to varying degrees, impede the widespread adoption of Joule heating recycling. Thus, in the current landscape, as opportunities arise, challenges also emerge.

5.1. Improving direct regeneration

5.1.1. Undamaged microstructure

The high temperatures provided by Joule heating may facilitate the structural repair of spent cathode and anode materials much better, with the objective of achieving an orderly crystal lattice and uniform elemental replenishment.

5.1.2. Electrochemical performance

The acceptance of recycled products is a controversial topic within the industry, representing a significant obstacle to the sustainable recycling of LIBs throughout their life cycle. One potential solution to this challenge is the restoration of the electrochemical properties of recycled materials, which could be better with the Joule heating strategy.

5.2. Innovating further upcycling

5.2.1. Surface engineering

Surface engineering is a common method of optimizing materials which can be used to improve certain aspects of recycled material properties (including rate, ICE cycle, etc.) while meeting the flash recycling of spent LIBs.

5.2.2. Doping modification

One of the most used modification methods for battery materials is doping, which can bring new fate to trace impurity elements remaining in spent LIBs. The objective is to enhance the electrochemical performance of the materials by replacing some of the anions/cations with doping elements with the adaptation of the atomic space architecture.

5.3. Inspiring industrial recycling

5.3.1. Universality of crafts

Joule heating recycling is built on the established traditional recycling process, offering a promising solution for efficiently processing a diverse range of LIBs waste materials in two distinct pathways (including NCM, LFP, graphite etc.).

5.3.2. Balance between input and output

Considering the unique characteristics of the two Joule heating recycling pathways, it is essential to reassess the energy consumption of different heating means and the carbon footprint of plant operations to align with the economic demands of large-scale production in the future.

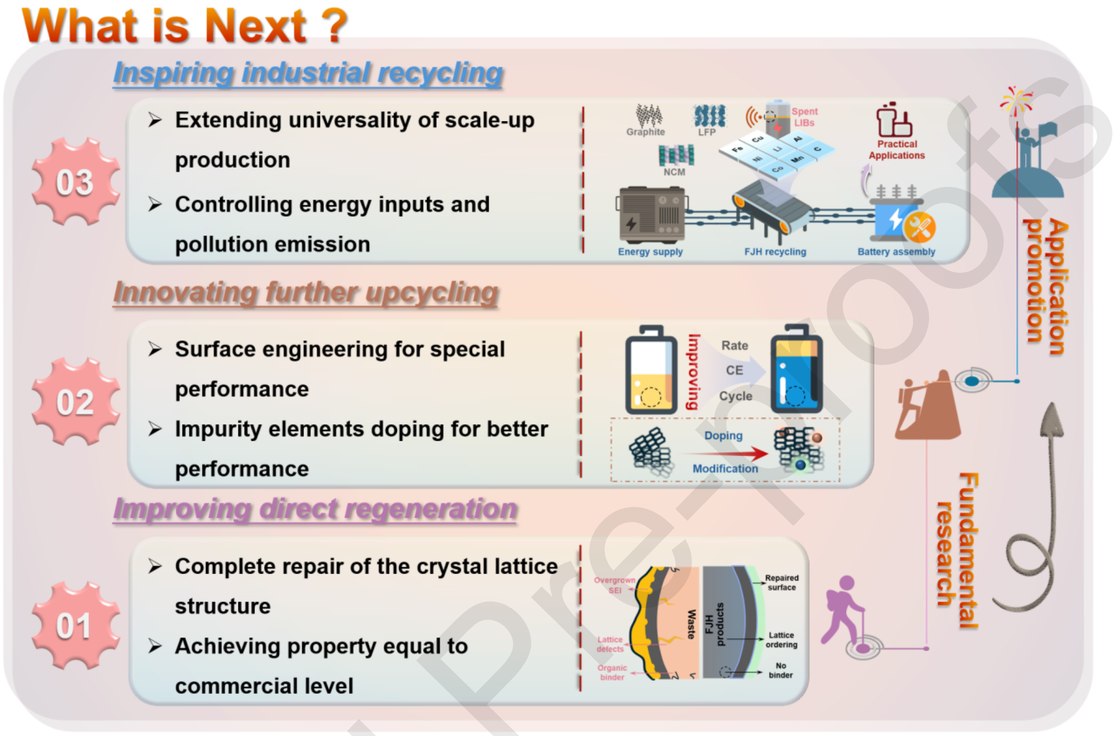

Fig. 6. Challenges and prospects of Joule heating recycling of spent LIBs.

In recent years, Joule heat, as a means of electrothermal synthesis, has been employed in the recycling of LIBs. Rapid LIBs recycling is accelerated by this method, whether the heating strategy is contact or contactless. However, further research is required on this technology for direct recycling of LIBs, particularly in terms of basic application and future promotion. Achieving a balance among product performance, recognition, and economic benefits of recycling is essential. The foundation for this research is rooted in the established principles of direct regeneration, thus necessitating the exploration of novel approaches to Joule heating recycling, including material upgrading, impurity utilization, and universality expansion. Moreover, advancements and innovations in joule heat equipment are pivotal factors contributing to its utilisation in rapid recycling application of spent LIBs. These innovations are poised to elevate Joule heating recycling to new heights and may propose overhaul of conventional lengthy and high-cost recycling process.

CRediT authorship contribution statement

Haoxuan Yu: Writing–original draft, Writing–review & editing, Investigation. **Meiting Huang:** Writing–review & editing. **Yifeng Li:** Writing–review & editing. **Liang Chen:** Writing–review & editing, Funding acquisition. **Hui Lv:** Funding acquisition. **Liming Yang:** Funding acquisition, Visualization. **Xubiao Luo:** Funding acquisition.

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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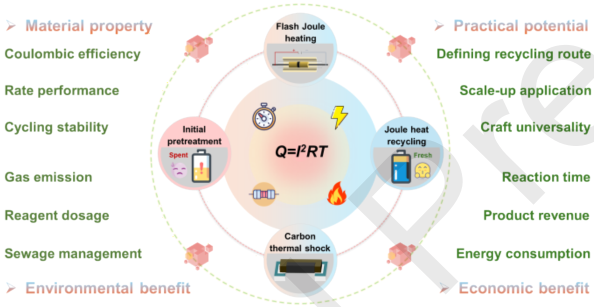
Graphical Abstract

The focus of this text is Joule heating recycling, represented by both flash Joule heating and carbon thermal shock, with the aim of comprehensively considering the advantages of aspects of flash recycling of spent LIBs.



Declaration of Interest Statement

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.