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Recycling cathode materials for lithium-ion batteries via Hydro-to-Cathode[®] method

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Due to the rising price and limited resource supply chain of Li [Ni_xMn_yCo_z]O₂ (x + y + z = 1) (NMC) cathode material, lithium-ion battery (LIB) recycling technologies have been emerging as the best solution to address the price issue. Mainly, conventional hydrometallurgy processes have been applied to the LIB recycling field in recognition of its value. One remarkable advantage of the hydrometallurgy method is that it serves as a bridge to enable the Hydro-to-Cathode[®] method. However, using recycled raw materials in the production of precursor cathode materials needs to be studied in parallel with the impurity (dopant) effect. The insufficient selective impurity removal technology leads to unexpected electrochemical properties in the final NMC cathode active material, which can be doped by several different impurities. Consequently, scrutinizing dopant elements (inorganic and organic) is critical if we want to consider the Hydro-to-Cathode[®] method as a major recycling process of NMC cathode material.

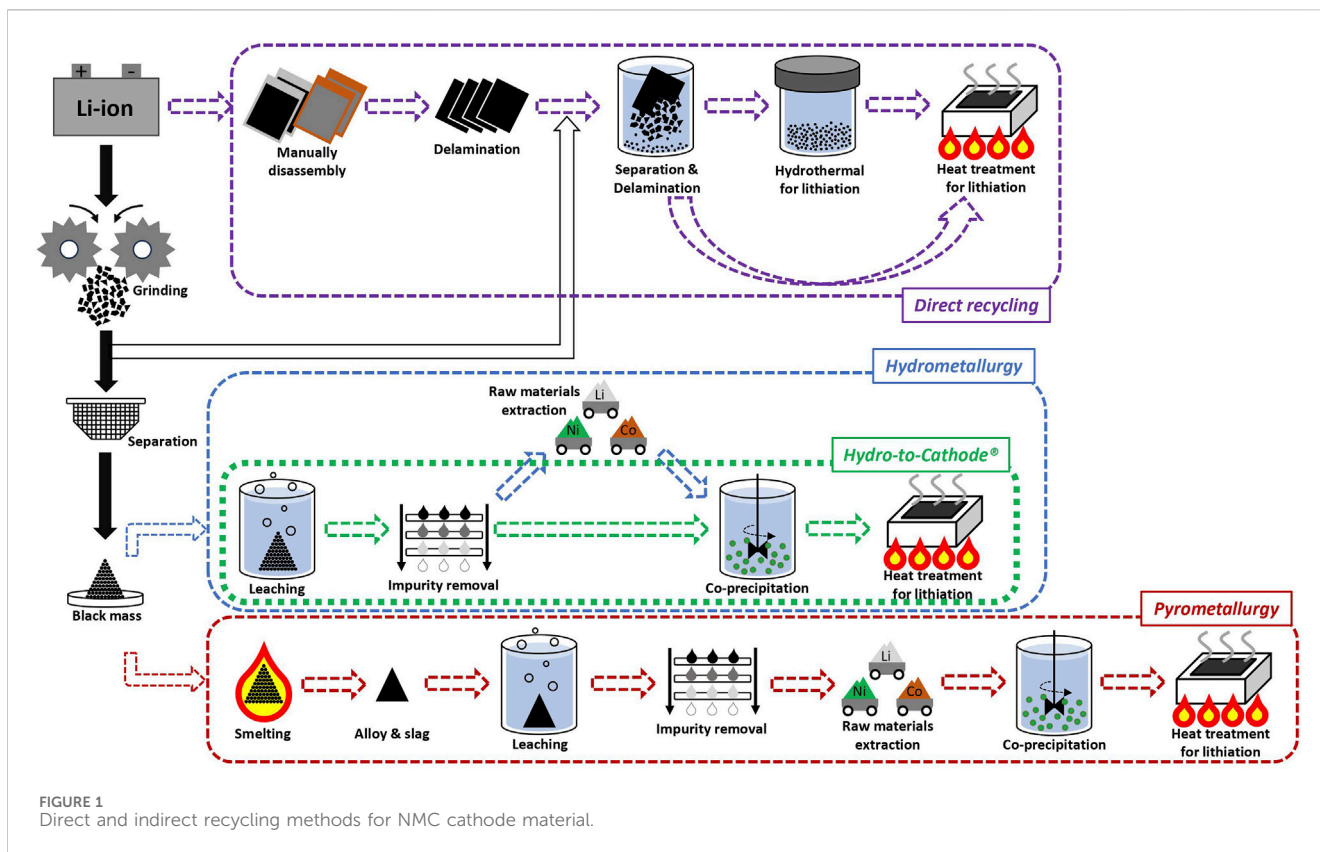
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recycling, lithium-ion battery, Hydro-to-Cathode[®], cathode material, closed-loop system

1 Introduction

Since the concept of the lithium-ion battery (LIB) was formalized by John Goodenough, Stanley Wittingham and Akira Yoshino, the LIB was broadly applied to a variety of industries such as electronic devices, military applications, and electric vehicles (EVs) (Bai et al., 2020b; Kim, 2022; Sim et al., 2023). More specifically, the market for EVs has dramatically increased; forecasting 140 million EVs to be operated worldwide by 2030 (Chemical and Engineering News (2019)). When compared to conventional combustion engine vehicles, EVs have relatively simple mechanical systems. They also generate low outgas emissions, resulting in greater attention relative to lower environmental impact.

Despite those advantages, the initial version of EVs has been challenging because of limited driving distance per single charge of the LIB. Following the first designed cathode material produced for commercial use, the ternary system of Li [Ni_xMn_yCo_z]O₂ (x + y + z = 1) (NMC) LIB cathode material has been in the spotlight. High Ni NMC based LIB cathode material [Li(Ni_xMn_yCo_{1-x-y})O₂, x ≥ 0.8] demonstrated particularly high energy density (200–250 mA h/g) and high operating voltage (≥4.3 V vs. Li⁺/Li) compared to LiFePO₄ (LFP), LiMn₂O₄ (LMO), and LiCoO₂ (LCO) based cathode materials, making it sufficient to meet the growing demand of the EV market (Li W. et al., 2020). That said, the high Ni NMC system demonstrates relatively insufficient cycling performance due to poor structural stability (Duan et al., 2019). To compensate



for this limitation, researchers have been studying techniques for doping, coating, and tailoring crystalline structure by introducing different elements and solid-state reaction skills (Ko et al., 2023; Susai et al., 2023; Tang et al., 2023; Hong et al., 2024).

Nevertheless, the LIB must be replaced every 5–10 years, at which it reaches end of life (EOL) due to the degradation of internal parts and materials (Mishra et al., 2022). Consequently, permanent use of the LIB is mostly impossible. The serious degradation of the cathode active material over time is a major cause of the reduced life of LIB. Cathode materials are an essential part of LIB, as they not only can determine performance of EVs, but also constitute more than half of the entire manufacturing cost of the LIB (Houache et al., 2022).

Consequently, due to the high demand of the LIB cathode materials, over time the prices of the LIB raw materials have lacked both control and consistency globally. Li is an essential raw material for cathode material as a source of electrochemical potential compared to others and has been designated as a strategic material for all countries (Bae and Kim, 2021). Ni and Co are also important elements, in terms of energy density and structural stability respectively (Ye et al., 2021). Therefore, many countries have focused on securing these materials. As a result, the price of Li, Ni, and Co have been rising recently. To overcome high price concerns, recycling technologies for LIB cathode materials are considered more commonly in both academic and industrial applications (Jung et al., 2021). In this review, we will cover several recycling methods and subsequent obstacles during the recycling process.

2 Recycling methods

2.1 Direct recycling

In terms of LIB recycling, there are two substantive approaches to accomplish the demanding task. The first, direct recycling, is an affordable method to meet goals such as cost reduction and suppressing environmental crisis. Compared to indirect recycling methods (pyrometallurgy and hydrometallurgy), direct recycling is more advantageous in terms of simplicity, efficiency, and conservation of energy. It can be broken down into three steps including grinding/delamination, separation, and regeneration (Figure 1). After collecting EOL LIB, they go directly through the grinding process or manual disassembly. From there, cathode material must be separated from grinded powder or delaminated film, which may contain anode, current collectors, and separator. Finally, the refined cathode material would be regenerated through proper treatments (Bai et al., 2020a; Xu et al., 2020; Kim et al., 2021).

When it comes to the regeneration process, it requires re-lithiation treatment via heat treatment because the most common failure of cathode material is attributed to Li loss during processing. Typically, the heat treatment process can be proceeded by using Li_2CO_3 as a Li source and has been widely studied due to its high accessibility (Zhang et al., 2013; Ji et al., 2023). Mainly, the regeneration of LMO and LFP based cathode materials has been studied due to low material cost and simple chemical composition. As the cost of those materials is very inexpensive, the conventional recycling method is not needed in this case (Lv et al., 2018). Besides, LCO and NMC cathode materials have been considered for direct

recycling by using Li_2CO_3 as a Li source (Nie et al., 2015; Shi et al., 2018). For heat treatment, duration time and temperature should be the most critical factors in addition to the Li source as the primary influencing factor. Though Li_2CO_3 is commonly utilized for re-lithiation studies, LiOH and LiNO_3 sources were verified from little research. The LiOH source was selected for the hydrothermal re-lithiation process which follows heat treatment (Figure 1) (Sloop et al., 2020). The hydrothermal treated recycled cathode materials showed relatively high cell cycling performance (Xu et al., 2021). Interestingly, there is a way to dramatically reduce lithiation temperature. Huang et al. reported that degraded Li $[\text{Ni}_{0.6}\text{Mn}_{0.2}\text{Co}_{0.2}]\text{O}_2$ (NMC622) material was sintered with LiOH- LiNO_3 molten salt for not only re-lithiation but also conversion to single crystal particle to allow for a single body type of cathode material that demonstrated high cycling properties (Huang et al., 2022).

Nonetheless, the direct recycling method lacks the flexibility to obtain different cathode chemistries. For instance, the used Li $[\text{Ni}_{1/3}\text{Mn}_{1/3}\text{Co}_{1/3}]\text{O}_2$ (NMC111), Li $[\text{Ni}_{0.5}\text{Mn}_{0.3}\text{Co}_{0.2}]\text{O}_2$ (NMC532), and NMC622 chemistries could only be recycled to the exact same NMC chemistry. Therefore, direct recycling cannot satisfy the demand for high Ni NMC cathode materials (Neumann et al., 2022). Even if there is no disruption in the supply of used high Ni NMC, due to the lack of structural stability it is realistically difficult to adopt the direct recycling method for high Ni NMC (Gao et al., 2022). In addition, due to the nature of this recycling concept, electrode, or LIBs cell current collectors such as Cu and Al must be perfectly separated after crushing. It is also worth noting that achieving perfect separation between cathode material and polyvinylidene fluoride (PVDF)/carbon is very challenging considering current technologies (Zhan et al., 2020). In summary, the direct recycling method is not fully applicable to support scaling up to industrial levels as it lacks required technologies.

2.2 Indirect recycling

The second predominant recycling method, indirect recycling, is based on deconstruction through smelting and leaching, and is widely preferred in the industry. Therefore, the pyrometallurgy and hydrometallurgy methods are prevailing routes for the execution of recycling LIB cathode material (Figure 1) (Bae and Kim, 2021; Kader et al., 2021; Latini et al., 2022; Mishra et al., 2022). In terms of recycling process simplicity, pyrometallurgy offers greater simplicity compared to hydrometallurgy. Considering both powder containing pretreated precious metals (black mass) from pretreatments (grinding and separation) or disassembled LIB can directly go through the smelting process, pyrometallurgy offers a simple operation and shortened process time (Latini et al., 2022). However, the smelting process is not viable in terms of energy saving and environmental impact because of high energy consumption and carbon emissions, respectively (Kader et al., 2021). Furthermore, extraction of Li is challenging because it can be incorporated into slag (Makuza et al., 2021; Miao et al., 2022). To overcome the impediments of the pyrometallurgy method, pyrolysis, roasting, and calcination processes have been introduced, allowing relatively low temperature conditions and higher recovery rates of Li (~93%) and Co (~99%) (Makuza et al., 2021). Nevertheless, the

pyrometallurgy method always requires a leaching process to retrieve precious metals as a post treatment.

Accordingly, the hydrometallurgy method has been selected for many years in the LIB cathode material recycling industry. The hydrometallurgy approach is based upon dissolution of spent LIBs via high acidic treatment and extract processes (Lv et al., 2018; Kader et al., 2021; Latini et al., 2022). The entire process involves multiple steps. During acidic treatment, unpleasant gas emissions and a significant volume of wastewater generation are unavoidable (Latini et al., 2022). However, the hydrometallurgy method can achieve high recovery yield as well as high purity of recycled metals. Particularly, the hydrometallurgy method encompasses several different processes such as pretreatment of the battery cell, leaching by acidic solution, selective precipitation of metal, impurity removal, and solvent extraction for drawing Li, Co, Ni, and Mn, separately. Among these processes, the leaching process has been actively studied - not only is the rate of recovery efficiency directly related to the high final yield of precious metals, but it is also directly linked to environmental pollution due to the generation of tons of wasted water after the process (Jung et al., 2021). Naturally, many researchers have been searching for highly efficient and minimally toxic leaching agents. Organic acids are a remarkable leaching agent in terms of environmental benefit. However, the high hurdle for adopting them at industrial scale has deterred widespread use. Therefore, inorganic acids have been broadly utilized for industrial purposes (Or et al., 2020). More specifically, the H_2SO_4 leaching system is widely studied for various methodologies of process through diversification. The H_2SO_4 system has relatively high adaptability to industrial scale compared to other inorganic acids such as HCl, HNO_3 , and H_3PO_4 . Therefore, different concentrations of H_2SO_4 , leaching temperature, additives, etc. can be controlled in terms of different leaching efficiencies for targeted precious LIB materials (Li, Ni, and Co) (Or et al., 2020; Chan et al., 2021; Jung et al., 2021).

2.3 Hydro-to-Cathode[®] method

The H_2SO_4 leaching system can also allow direct resynthesizing via a co-precipitation process using the purified leached metal solution (Figure 1). This concept was originally patented by Yan Wang et al. in United States, and they named it the Hydro-to-Cathode[®] method for the very first time (Wang et al., 2013). The Hydro-to-Cathode[®] method is advantageous for both cost reduction and precursor cathode materials design. The concept itself is designed for creating a closed-loop system, which provides a better solution than conventional recycling methods to improve process time and cost in the LIB material industry. Therefore, research on the Hydro-to-Cathode[®] concept is actively being conducted in both commercial and academic fields. Ascend Elements plans to open a new large scale LIB precursor cathode and cathode materials plant in the United States using the Hydro-to-Cathode[®] method (Recycling Today, 2022). Compared to the direct recycling approach, the Hydro-to-Cathode[®] method not only allows increased accessibility to various cathode chemistries, but also tailoring crystalline structure and surface morphology of precursor cathode material for certain models of battery cells. The Yan Wang group conducted a preliminary study to verify the potential of the Hydro-to-Cathode[®] method. They showed a

direct comparison between recycled and virgin raw NMC metal solutions by a co-precipitation process to evaluate final cathode materials.

The Yan Wang group then went through a leaching process with mixed cathode materials [LCO, LMO, NMC111, and LFP]. During this process, Fe was properly removed by pH adjustment and Li_2CO_3 was recovered. After that, $\text{Ni}_{1/3}\text{Mn}_{1/3}\text{Co}_{1/3}(\text{OH})_2$ was synthesized by a recycled metal solution. Furthermore, final cathode material was prepared by using the recovered Li_2CO_3 . The recycled cathode material showed a similar range to commercial grade of electrochemical performance. They have also reported recycled cathode material is more economical compared to the normal process. (Zou et al., 2013).

Beyond academic field study, they recycled actual LIBs for getting final NMC111 product. Before the leaching process, pretreatments (shredding, magnetic removal, aluminum dissolution, and sieving) were systematically proceeded. For characterization, the final product was analyzed by X-ray diffraction (XRD) measurement, and the peaks matched well with layered structure without any abnormal impurity peaks (Gratz et al., 2014).

They also reported that four different feedstocks of EOL LIBs from various manufacturers were utilized for the Hydro-to-Cathode[®] method. Four precursor cathode materials were co-precipitated to obtain $\text{Ni}_{1/3}\text{Mn}_{1/3}\text{Co}_{1/3}(\text{OH})_2$. The final cathode materials were evaluated with coin cell testing to characterize their electrochemical properties. All four samples showed similar rate capability to commercial cathode material at low C-rates, while they exhibited superior capability in high C-rates. Therefore, they could conclude that the Hydro-to-Cathode[®] method has minimal impact on the final recycled cathode material (Zheng et al., 2018).

Lastly, they executed one batch experiment with up to 30 kg of spent LIBs from different EV manufacturers. The recycled cathode material was compared with commercial cathode material by both coin cell and single layer pouch cell tests. Recycled and commercial cathode materials showed similar properties through discharge ΔSOC (state of charge) tests and direct current resistance tests. This was the first trial for the scaling-up process, so they could confirm Hydro-to-Cathode[®] is a suitable method in terms of scalability (Chen et al., 2019).

Other groups further investigated the effects of impurities in final cathode material. Beak et al. verified that actual recycled NMC metal solution can affect morphology control of precursor cathode material $[\text{Ni}_{1/3}\text{Mn}_{1/3}\text{Co}_{1/3}(\text{OH})_2]$. They reported after converting to cathode material, high levels of metallic impurities could have an unfavorable impact on initial charge/discharge capacity and capacity retention at 55 cycles at coin half-cell test (3.0–4.3 V). They also argued that metallic and nonmetallic impurities are advantageous as dopants in cathode material to get better rate capability than non-doped cathode material (Beak et al., 2022).

Kauppinen et al. introduced recycled and purified Mn for Li $[\text{Ni}_{0.8}\text{Mn}_{0.1}\text{Co}_{0.1}]\text{O}_2$ (NMC811) cathode material. They synthesized precursor cathode materials with three different metal sulfate solutions which contain different levels of metallic impurities (Zn, Ca, K, Mg, and Fe). Following proper sintering processes, they assessed the electrochemical property through pouch cell testing (2.8–4.2 V). After 1,100 cycles, capacity retention showed over 80% and initial discharge capacity reached 184 mA h/g.

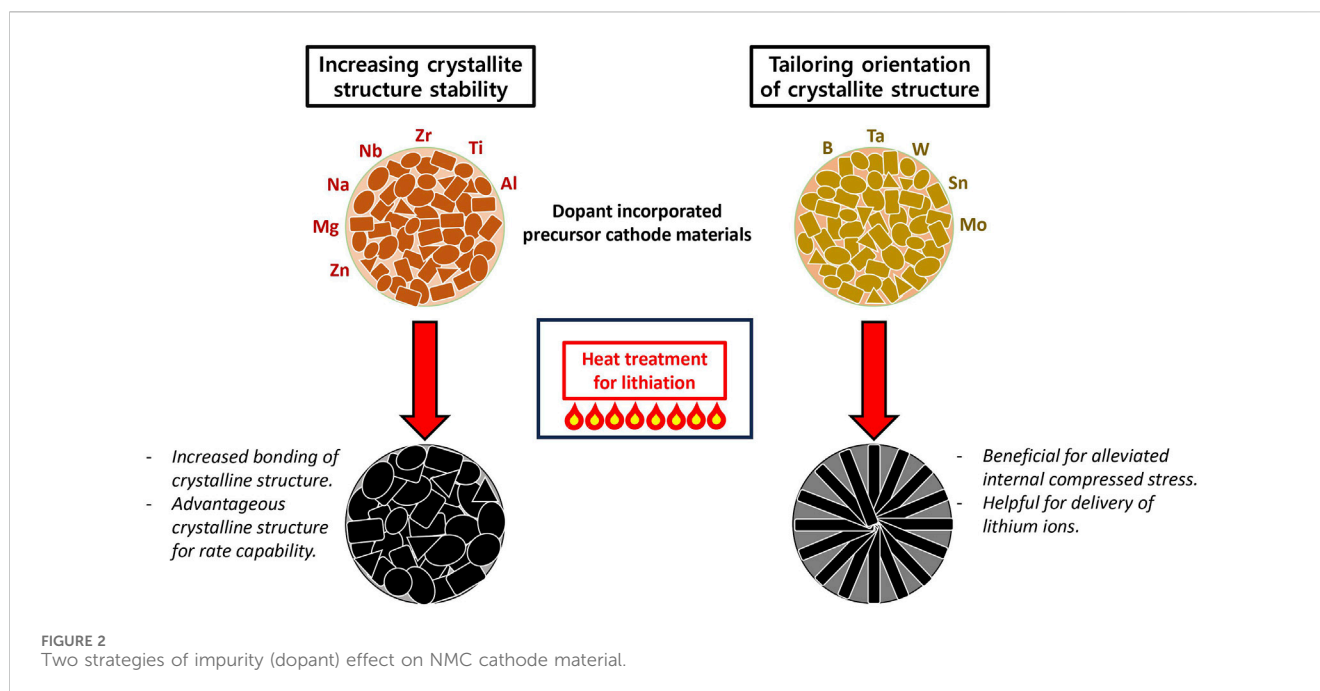
Therefore, they concluded the recycled raw material is feasible for high Ni NMC cathode material (Kauppinen et al., 2023).

2.4 Impurity (dopant) control for the Hydro-to-Cathode[®] method

The mentioned experiments with NMC based cathode materials can be considered important because they have studied the real possibilities of the Hydro-to-Cathode[®] method. The types and levels of impurities would not be easily controllable by impurity removal during the recycling process. Even a small ppm level of impurities could have severe influence on electrochemical properties as a dopant. Furthermore, the combination of impurities with two or three elements can provide different aspects of cell performance rather than what we expected (Tian et al., 2023). Therefore, scrutinizing the doping effect on cathode materials is indispensable if we rely on precursor NMC cathode material synthesized via co-precipitation with recycled metal raw materials.

In the last decade, doping effects of specific elements have been investigated through NMC cathode material. In accordance with the operation mechanism of doping in the NMC system, we can divide into two main doping strategies (Figure 2). The first scheme involves introducing similar size of element to either Li or transition metal atoms. These specific elements make it possible to substitute either transition metal atoms or Li. Among all possible elements, Al, Ti, Zr, and Nb are especially considered as structural stabilizers due to their high bonding with the neighbor transition metal atoms (Schipper et al., 2016; Liu et al., 2018; Song et al., 2018; Xin et al., 2019; 2022; Yang et al., 2019; Zhang et al., 2019; Park et al., 2020; Zhou et al., 2021; Bizzotto et al., 2023). Therefore, they can lead to high cycling performance and capable performance under high voltage conditions. Other feasible dopants are Na, Mg, and Zn, which can play a pivotal role in improving rate capability by replacing the Li site as a pillar (Du et al., 2015; Huang et al., 2016; Li et al., 2019; Li L. et al., 2020; Liu et al., 2019; Sattar et al., 2020; Gomez-Martin et al., 2022).

The second strategy involves tailoring a preferentially aligned NMC cathode crystalline structure, rather than chemically bonding with a heterogeneous element and the main NMC structure to mitigate collapse of cathode material crystalline structure. For instance, a radial columnar structure, which is designed to extend outward from the center, could be advantageous for getting enhanced electrochemical performance. This uniquely oriented crystalline structure can diminish internal stress and strain caused by charge and discharge, allowing dramatically improved cycling performance to be achievable. B, Ta, W, Sn, and Mo have been studied for forming controlled radial columnar structure by many researchers (Park et al., 2018; Lee et al., 2019; Ryu et al., 2019; Kim et al., 2020; Sun et al., 2021; Thien Nguyen et al., 2021). In particular, B has been acknowledged as a key dopant in terms of structural controlling of NMC cathode material. Because the B doped cathode materials exhibited enhanced cycling performance and discharge capacity, it can be attributed to alleviated internal compressed stress and increased delivery of lithium ions during the charging and discharging processes. Ryu et al. conducted a direct comparison between NMC and Li $[\text{Ni}_x\text{Co}_y\text{B}_{1-x-y}]\text{O}_2$ (NCB) systems to observe B doping effect undoubtedly. They also demonstrated that different doping amounts and sintering temperature could be



an adjustable factor to find optimum crystalline structure (Ryu et al., 2020).

3 Discussion and future works

In this mini review, we have discussed several recycling methods for obtaining precious materials from the EOL LIB. It can be divided into two broad categories including direct and indirect recycling methods. The direct recycling method is an attractive approach in terms of saving cost, time, and energy due to the simple nature of the process. However, its technical challenges and limited freedom of NMC composition will limit its utilization in the commercial LIB industry. In contrast, the indirect recycling method provided relatively clear direction toward industrial application. The indirect recycling method is approachable through pyrometallurgy or hydrometallurgy processes. Technically, these processes require completely different paths to break down into precious major raw materials such as Li, Ni, and Co. However, both methods mandatorily go through leaching and extraction processes to draw the raw materials. Therefore, the hydrometallurgy method is advantageous for industrial application. Moreover, the Hydro-to-Cathode[®] method, which is an advanced variation of the hydrometallurgy method, showed positive signs to attain a closed-loop system. The remaining ppm level of impurities can be doped into the precursor cathode materials, so that the final cathode material will exhibit significantly improved electrochemical properties compared to the virgin cathode material. The improved performance of the cathode material can be attributed to two doping strategies including strengthened structure and oriented crystallite structure. Consequently, the Hydro-to-Cathode[®] method would be successfully adopted into the industrial field with a genuine understanding of dopant (impurity) impact and precise adjustment skill for impurity control.

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Conflict of interest

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