

Grand Challenge

Grand challenges in recovery of critical elements from end-of-life lithium-ion batteries

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Lithium-ion batteries (LIBs) are widely used for various applications that vary from consumer electronics to national defence in the field of energy storage and batteries. In recent years, hybrid and electric vehicles have become popular owing to which the demand for LIBs has grown exponentially. The elements such as cobalt (Co), nickel (Ni), lithium (Li), and manganese (Mn) are critical components in LIB manufacturing. Moreover, the chemistry and composition of LIB materials vary widely based on applications. The purity requirements of critical materials in the LIB cathodes are >99.9 wt.%. Thus, there is a critical need of high purity battery grade materials, and it is forecasted to increase very significantly. However, the global resources of these materials are limited and are declining rapidly. On the other hand, the widespread applications of LIBs are expected to generate millions of end-of-life LIBs in future years. Considering the growing demand of high purity battery grade materials, the scrap LIBs could be potential source of the critical battery materials. However, the LIB industry lacks a clear path to large scale recycling due to several challenges involved in the recovery of critical materials from end-of-life LIBs. These challenges and potential solutions on those challenges are discussed in this review.

KEYWORDS: Lithium-ion batteries; LIBs; Recovery; Critical elements.

GRAPHICAL ABSTRACT**HIGHLIGHTS**

- Rising lithium-ion battery demand has exponentially strained global resources of critical materials.
- Meanwhile, growing numbers of end-of-life LIBs could serve as a source for recovering critical materials.
- Various challenges hinder LIBs recycling, including process complexities, cost, and efficiency issues.
- Cost-effective, energy-efficient, and green technologies, scaled and globally adopted, can address battery material recovery challenges.

1. Introduction

Since the first commercialization efforts approximately 30 years ago, lithium-ion batteries (LIB) have attracted worldwide attention owing to their versatility, and wide use in consumer electronics, portable appliances, hybrid and electric vehicles (Sun, et al., 2018). The total energy per unit volume provided by LIBs is significantly higher than the conventional nickel-cadmium (Ni-Cd) or nickel-metal hydride (NiMH), and lead-acid batteries (Li, et al.,

2010). Some other advantages of LIBs include lighter weight, no memory effect, and a slow self-discharge rate.

LIBs contain several critical metals such as Co, Li, Ni, and Mn as well as graphite which is widely used in LIB anodes. Co is a critical material, substantially and increasingly used in the lithium-ion batteries (LIBs). Currently, the terrestrial cobalt resources in the world are estimated at

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approximately 25 million tons, whereas the accumulated cobalt reserves in the world are approximately 7.6 million tons (Shedd, United States Geological Survey (USGS), 2022). According to U.S. Geological Survey (USGS), 170,000 tons of cobalt was produced through mining and other activities. The richest sources of cobalt are primarily in the Democratic Republic of Congo which currently supply more than 70% of the global cobalt demand (Shedd, United States Geological Survey (USGS), 2022). Additionally, China, Russia, Australia, and Canada contribute to the global supply to some extent (Chen, et al., 2019, Zubi, et al., 2018). Australia and Chile are the leading contributors in global Li production through mines. Due to continuous explorations and increased applications for Li, the identified global Li resources have increased substantially to 89 million tons, whereas the global reserves are approximately 22 million tons. Bolivia has the largest Li resources contributing to 23% of the global Li resources (Jaskula, United States Geological Survey (USGS), 2022). Mn is mainly extracted from terrestrial sources in South Africa, Gabon, and China. Although, land-based resources of Mn are irregularly distributed and have very low purity which leads to high extraction costs. Currently, the global Mn reserves are approximately 1.5 million tons (Schnebele, United States Geological Survey (USGS), 2022). Finally, global Ni production is dominated by Indonesia, and Philippines while Canada, Russia, and Australia contribute to some extent (Steward, et al., 2019). The total Ni reserves are more than 95 million tons. Approximately 2.7 million tons of the Ni reserves are produced through mines (McRae, United States Geological Survey (USGS), 2022). The leading countries in graphite production are China, and Australia. China contributed to approximately 79% of the 1.2 million tons of natural graphite produced through mining. The total graphite reserves in the world are approximately 320 million tons whereas the total global resources exceed 800 million tons (Olson, United States Geological Survey (USGS), 2022).

The LIB industry is projected to consume approximately more than 60% of the global cobalt reserve by 2040 and the LIB demand is expected to outgrow the raw materials supply by 2030 (Alipanah, et al., 2023). Hence, LIBs are considered as an important secondary resource for the extraction and recovery of cobalt (Golmohammadzadeh, et al., 2018). Within the next few years, the global critical materials reserves will be under pressure to fulfil the growing demands of LIB industry. Thus, there is a critical need for high purity battery grade materials which is forecasted to increase very significantly. The analysts

have predicted a 575% increase in Li demand and 1237% increase in Ni demand for LIB production in the next 10 years (Beaudet, et al., 2020, Bruder Müller, et al., World Economic Forum: Global Battery Alliance, 2019). However, the global resources of these materials are limited and are rapidly declining (Golmohammadzadeh, et al., 2018, Jagannath, et al., 2017). Therefore, the end-of-life LIBs should be properly handled and recycled to effectively use the limited resources of critical materials. Furthermore, the waste generated from spent LIBs contain large quantities of metallic contaminations which affect the environment adversely. Co and Ni are classified as carcinogenic, and mutagenetic materials whereas the organic electrolytes used in LIBs are detrimental to human health and environment (Fan, et al., 2020). Considering these factors, the LIB recycling is one of the most pursued research areas in the past decade. Figure 1 shows the growing trend in the patents published within last 2 decades. Figure 2 provides the geographical distribution of research organizations and companies active in LIB recycling research and patents filed in the last 5 years in the recycling of LIBs and critical materials in LIBs.

On the other hand, the ever-growing applications of LIBs in recent years in various industries are expected to generate millions of tons of end-of-life (EOL) LIBs in future years. Over 5 million metric tons of LIBs are expected to reach EOL by 2030 which has given rise to serious environmental concerns. Considering the growing demand for high purity battery grade materials, the scrap or spent LIBs are potentially a secondary source of the critical battery materials with cost-effective recovery and recycling. However, some major challenges in LIB recycling can create obstacles in providing an adequate supply of critical materials. Some of these challenges include the chemistry and composition of LIB cathodes, complexity of the spent LIB feedstock, purity requirements of raw materials for LIB fabrication, the chemical forms of these raw materials (sulfates, carbonates, hydroxides, etc.), and several other limitations of the current recycling technologies (Castillo, et al., 2002, Chen, et al., 2019, Hanisch, et al., 2015, Ku, et al., 2016, Lai, et al., 2021, Ma, et al., 2021, Mroziak, et al., 2021, Wei, et al., 2023, Zubi, et al., 2018). Therefore, there are only a few commercialization efforts around the globe related to recycling of LIBs. In this review, various approaches followed for the recycling of LIBs, grand challenges in LIB recycling, and future perspectives will be discussed.

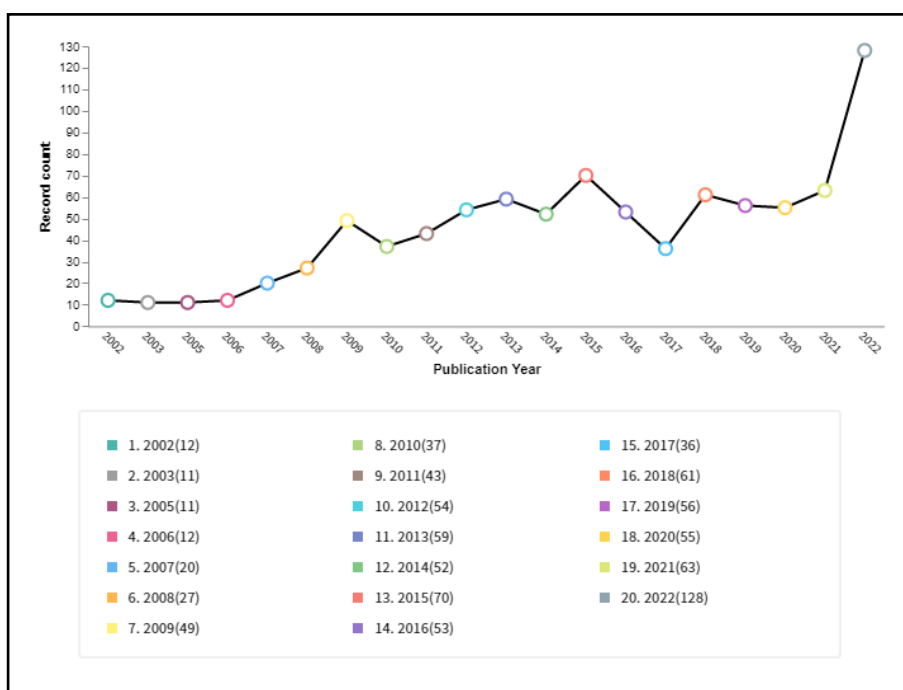


Fig. 1. Growing trend of patents published in the research field of LIB recycling.

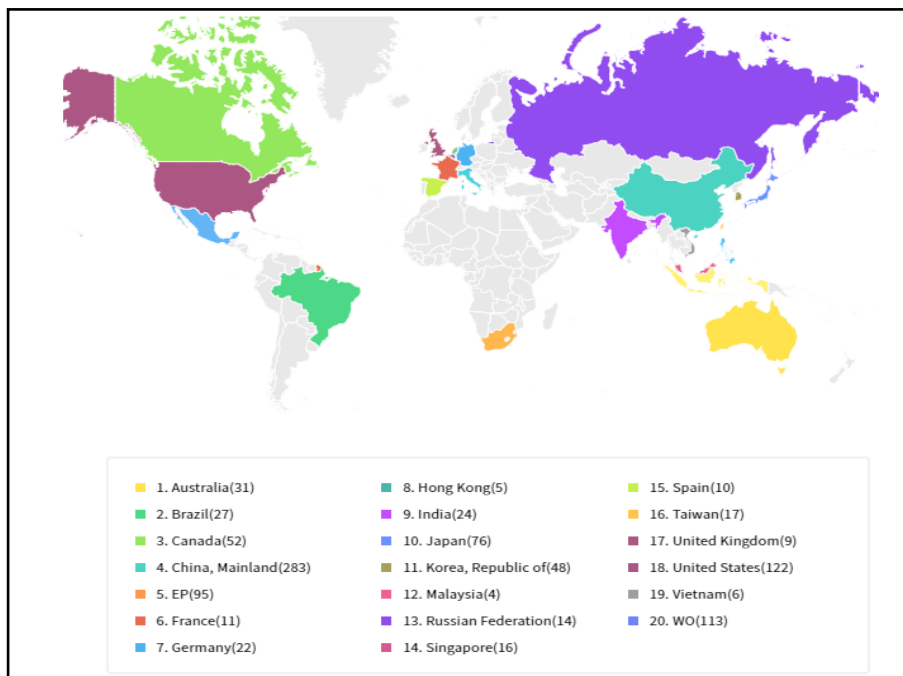


Fig. 2. Geographical distribution of patents filed in the last 5 years in the LIB recycling research.

2. Review scope and approach

This review focuses on recycling of LIBs and grand challenges in various LIB recycling approaches in addition to the outlook into LIB recycling industry. Various online databases such as PubMed, Science Direct, Google Scholar, Knovel, SciFinder, and Derwent Innovation were used to conduct the review. The keywords “spent lithium-ion battery”, “recycling of spent LIBs”, “challenges in LIB recycling”, “waste management in LIB recycling”, “Perspectives in LIB recycling”, “separation and recovery technologies”, “recovery and recycling of critical materials”, “metal recovery”, “hydrometallurgy, solvent extraction, and pyrometallurgy”, “membrane separations for LIB recycling” etc. were used to narrow down the search. The articles were then checked for quality, topicality, and relevance based on the journal’s impact factor, the year of publication, and citations of the articles.

3. LIB composition

LIB packs are composed of a cathode, an anode, and organic electrolyte and a polymer-based separator. These components are laminated and compressed together to create an electrical contact between them (Ordoñez, et al., 2016, Steward, et al., 2019). Cathode materials generally consist of combinations of active metal powders including Co, Ni, Mn, Al, Fe, etc. depending on the application and desired electrical quantities. Some of the typical cathode material combinations are LiCoO_2 , $\text{LiNi}_x\text{Mn}_y\text{Co}_z\text{O}_2$ ($\text{Ni}:\text{Mn}:\text{Co} = 1:1:1, 6:2:2,$ or $8:1:1$), Lithium manganese oxide (LMO), Lithium iron phosphate (LFP), and Lithium nickel cobalt aluminum oxide (NCA). Natural and artificial graphite are generally used as common anode materials whereas separators are usually made from polymers. An organic electrolyte is used to submerge electrodes and acts as an inert component. Longevity, and stability against both the cathodes and anode materials are some of the desired characteristics in electrolytes and separators (Steward, et al., 2019).

4. LIB recycling approaches

In this section, various technologies and approaches followed in LIB recycling will be introduced and discussed briefly. Echelon or secondary

utilization and active constituent recovery after battery disassembly are primarily used approaches for the recycling (Wei, et al., 2023). LIBs are discarded from their use when the energy densities are dropped to approximately 80% of their original capacity. An efficient recycling strategy for LIBs is echelon utilization until the battery capacity decays to 40% followed by active constituent recovery (Castillo, et al., 2002, Lu, et al., 2022, Sathre, et al., 2015).

4.1. Echelon utilization

Echelon utilization is primarily applied to used/spent LIBs in less-stressful applications including smart grids and storage systems. It avoids the large-scale scrapping, maximizes the battery lifespan, and makes LIB recycling safer. In this phase of LIB recycling, batteries are sorted according to internal resistance, side reaction, residual life, and remaining capacity. Based on the sorting results, batteries are dismantled and reconfigured for echelon utilization (Lai, et al., 2021).

4.2. Active constituent extraction and recovery

After the secondary utilization, the batteries need to be recycled using material extraction to achieve circular economy. The extraction includes pretreatment, leaching of critical materials from waste LIB feedstock, separation, and purification of these critical materials.

4.2.1. Pretreatment

Pretreatment processes are applied in LIB recycling to separate materials that can be easily removed from battery packs. These materials are typically encapsulated in iron and plastic. Several treatments including mechanical separation, thermal treatment, mechanochemical treatment, and dissolution are applied in pretreatments. Mechanical separation consists of crushing, grinding, gravity, and magnetic separation. This coarse separation makes it easier to further treat smaller components in complex structures that cannot be easily

removed (Al-Thyabat, et al., 2013, Ku, et al., 2016). Thermal treatments are carried out to pyrolyze organic additives and binders so that active cathode and anode materials can be separated from the binders. This is also done to avoid any interference of binders or electrolytes in the recovery of individual critical materials (Hanisch, et al., 2015, Wei, et al., 2023). Mechanochemical treatment is used particularly for the decomposition of crystal structure of LiCoO_2 where Co and Li are extracted via an acid leaching process at room temperature (Saeki, et al., 2004). In some cases, dissolution through organic solvents is also utilized to dissolve binders. However, the high costs of organic solvents and equipment put some limitations on industrial adaptation of this technology (Sun, et al., 2018, Zeng, et al., 2015).

4.2.2. Metal extraction

Followed by pretreatments, critical materials including the metals in LIBs are leached using pyrometallurgy and hydrometallurgy. Pyrometallurgy involves combustion of pretreated LIBs in the smelter where the components are broken down with simultaneous decomposition of residual organic materials. The metals are converted to stable alloys in this process, which can then further be treated to obtain pure metals (Fan, et al., 2020, Paulino, et al., 2008). Pyrometallurgy is a simple and mature process that has been used widely. However, it has several disadvantages including efficiency, and waste generation which will be discussed in later section of the review. Hydrometallurgy consists of leaching metals into inorganic acids such as sulfuric acid, nitric acid, and hydrochloric acid. Typically, several process parameters such as high temperatures, sonication, and reducing agents are used to accelerate the leaching process (Lu, et al., 2022). In addition to inorganic acids, several organic acids such as citric acid, malic acid, aspartic acid, ascorbic acid are used due to their thermal stability, low environmental footprint, and recyclability (Lin, et al., 2021). Alkali and bioleaching have also been applied in hydrometallurgy to dissolve metals in spent LIBs (Chen, et al., 2018, Moazzam, et al., 2021). However, large consumption of water, corrosion, secondary pollution, slow kinetics, low leaching efficiency are some of the challenges that still remain in these novel extraction techniques (Golmohammadzadeh, et al., 2018, Lai, et al., 2021).

4.2.3. Metal element recovery

After dissolving the metals into the leaching solutions, individual critical materials or a combination of materials are separated and recovered using various techniques including solvent extraction, membrane-assisted solvent extraction, electrochemical separation, and precipitation. Solvent extraction is a well-known and widely used technology for the recovery of critical materials which takes advantage of the complex formation ability of metals and get distributed into two different phases with the help of an extractant. Numerous commercially available extractants such as bis (2,4,4-trimethyl-pentyl) phosphinic acid (Cyanex 272), di(2-ethylhexyl) phosphoric acid (D2EHPA), diethylhexyl phosphoric acid (DEHPA), dithiophosphinic acid (Cyanex 301) have been used successfully for the separation and recovery of critical materials from spent LIBs (Carson, et al., 2020, Santanilla, et al., 2021, Swain, et al., 2006, Swain, et al., 2015, Wang, et al., 2016). Chemical precipitation is another technique that leverages solubility of metal ions in the presence of OH^- , S^{2-} , $\text{C}_2\text{O}_4^{2-}$ to separate individual metals (Biswal, et al., 2018). Redox reactions have also been used for recovering metals from LIBs by applying a potential difference between anode and cathode (Armstrong, et al., 1996).

5. Challenges in spent LIB recycling

Although there have been significant advancements in fundamental and applied research in LIB recycling in the recent years, there are still many challenges and limitations to overcome in order to develop a recycling process of LIBs that is cost-effective, environmentally friendly, and energy efficient. Rapidly changing compositions, designs, and materials, waste generation, efficiency of the recycling technology, complexity of the spent LIB feedstock

make the recycling much more challenging. Currently, the LIB industry lacks a clear path to large scale recycling, since there are no commercially available technologies for LIB recycling. The state-of-the-art metal extraction technologies such as pyrometallurgy, hydrometallurgy, and solvent extraction that are primarily used in mining industries could be utilized in LIB recycling. However, these technologies have several limitations in terms of product purity, chemical usage, and process resilience for varying spent LIB feed compositions. The limitations as shown in Figure 3 will be discussed in detail in the following section.

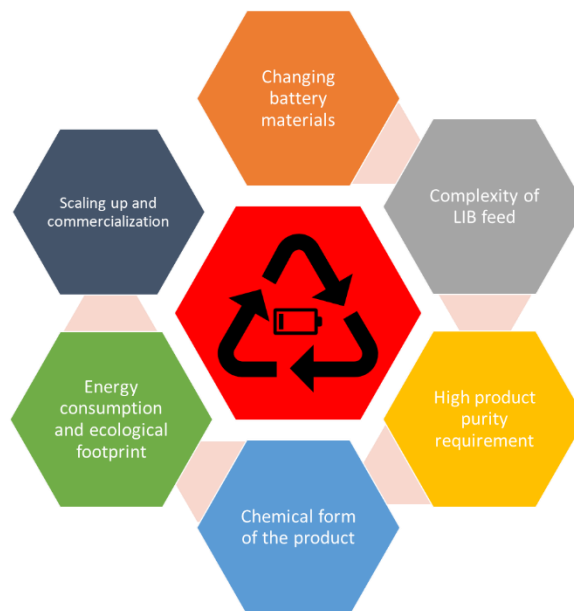


Fig. 3. Challenges associated in LIB recycling.

Table 1

Comparison between various types of LIB cathodes (Al-Thyabat, et al., 2013, Chen, et al., 2019, Hanisch, et al., 2015, Julien, et al., 2014, Manthiram, 2020, Murdock, et al., 2021, Saeki, et al., 2004, Sathre, et al., 2015, Zubi, et al., 2018).

Cathode	Crystal structure	Variants and compositions	Typical specific capacity (mAh/cm ³)
LiCoO ₂	Layered	LiCoO ₂	150
NMC	Layered	NMC 111	170
		NMC 532	
		NMC 811	
		NMC 622	
		NMC 955	
NCA	Layered	LiNi _{0.3} Co _{0.3} Al _{0.3} O ₂	200
LMO	Spinel	LiMn ₂ O ₄	100
LFP	Olivine	LiFePO ₄	160

5.1. Continuously changing battery materials

The materials and compositions of LIB cathodes vary widely based on LIB applications. Currently, layered oxides, spinel oxides, and polyanion oxides are the three main types of cathode materials that are in use. The most widely used LIB cathodes include lithium nickel manganese cobalt oxide (NMC such as NMC111, NMC532, NMC622, NMC811, and NMC955), lithium nickel cobalt aluminum oxide (NCA), lithium cobalt oxide (LCO), lithium manganese oxide (LMO) and lithium iron phosphate (LFP). Table 1 provides a detailed

comparison between various types of cathodes used in LIBs. However, due to the limited resources of critical materials including cobalt, there is a critical need to develop new cathode materials by adding a new metal in the composite oxide or introducing a new cathode composition altogether. Addition of new materials and/or modification in the cathode composition of LIBs pose new challenges in the technologies that are developed for established cathodes.

5.2. Complexity of the spent LIB feed (Black mass)

One of the major limitations in the recovery of critical materials from spent LIBs is the complexity of the spent LIB feed or “black mass”. In case of some

spent LIBs, the anodes, and cathodes from different types of LIBs are shredded together which results in a black mass of varied composition with both desirable elements such as Co, Mn, Ni, and Li, and undesirable elements such as Fe, Al, Cu, and Zn. The chemistries of LIB cathodes are constantly evolving due to supply constraints of critical materials and proprietary technology development by EV manufacturers. Therefore, there is a wide variability in black mass composition depending on the cathode chemistries. The traditional metal mining technologies may not be able to separate the critical materials in their pure form from the scrap LIBs since the scrap LIB feedstocks are much more complex than a typical mining feedstock.

Table 2

Comparison of LIB recycling technologies from energy and footprint standpoint (Boyden, et al., 2016, Du, et al., 2022, Rajaeifar, et al., 2021, Steward, et al., 2019, Velázquez-Martínez, et al., 2019, Velázquez-Martínez, et al., 2019)

Desirable attributes in LIB recycling technology	Existing LIB recycling technologies		
	Conventional Solvent Extraction	Pyrometallurgy	Hydrometallurgy
Single Step	Multi-step process	Multi-step	-
Low-cost off-the-shelf equipment	-	Specialized expensive equipment	
High purity			Low purity
High yield			Low yield
Minimal chemical usage	High chemical usage		High chemical usage
Minimal waste generation	Significant waste generation		
Low capital and operating cost	High capital and operating cost	High capital and operating cost	
No equilibrium limitation	Limited by equilibrium		
Ambient temperature operation		High temperature operation	

5.3. High purity and chemical form requirements

The minimum purity requirements of critical materials in the LIB cathodes are typically 99.9 wt%. High purity materials are required to ensure high performance of LIBs and extended battery life with a full current capacity. Due to the inefficient recycling processes, additional separation steps need to be applied to achieve the desired purity of recovered metals. This adds to the total cost of the recycling processes, making them less feasible economically. Furthermore, the materials are typically preferred by battery manufacturers in the form of sulfates which can be easily incorporated as raw materials in the LIB cathode chemistries to reduce the capital and operating cost of the process. The individual metals can be recovered in the form of oxides or oxalates using pyrometallurgy or hydrometallurgy. This gives rise to the necessity of a conversion step to convert oxides or oxalates to sulfates.

5.4. High energy consumption and ecological footprint

Existing LIB recycling processes such as pyrometallurgy and hydrometallurgy can mitigate the growing concern of environmental impacts of spent LIB waste. However, the inefficiencies in the operation of these techniques, generated waste, and emissions after the recycling processes leave a negative impact on the environment. For instance, the incineration required in pyrometallurgy leads to the release of toxic and greenhouse gases into the environment. Additionally, incomplete combustion may lead to harmful waste residue (Zheng, et al., 2018). Furthermore, pyrometallurgy is highly energy intensive which makes the technology a high footprint process. On the other hand, hydrometallurgy relies upon treatment of solutions after extraction or dissolution. This requires additional wastewater treatment, thereby increasing the ecological footprint. It can also pose environmental risks via contamination of freshwater sources (Mrozik, et al., 2021). Table 2 provides desirable qualitative attributes in an ideal LIB recycling technology and energy and footprint requirements in existing LIB recycling technologies.

5.5. Scaling up of the recycling technologies

Most of the technologies developed for LIB recycling have been demonstrated on bench scale and simplified, simulated feed solutions. In comparison, the industrial scale waste LIB feedstock generation is on a much larger scale and much more complicated. This mismatch between the academic research and industry conditions limits the commercialization of LIB recycling technologies. Additionally, the challenges mentioned above in this section reduce the profits margin thereby making them economically non-viable (Ma, et al., 2021).

6. Future perspectives

Growing demand and applications of LIBs lead to a vast number of end-of-life LIBs every year affecting the supply chain and the environment adversely. Although research advances and breakthroughs in the last two decades have addressed many challenges in LIB recycling, there is a critical need for cost-efficient, environmentally friendly, and energy-efficient processes. Additionally, it is imperative to follow the 3R strategy (reduce, reuse, and recycle) while using LIBs for various applications (Arshad, et al., 2020). Most LIB recycling technologies prioritize enhancing efficiency and purity of final products. While these two performance parameters are critical for the success of the recycling technology, it is important to consider the waste generation caused due to the technology. More attention needs to be paid to control and/or minimize the generation of waste and use of environmentally friendly reagents. Currently, many of the developed countries have policies in place and there is a push towards development of sustainable LIB recycling. However, the awareness should be raised amongst citizens since they are the primary users. Additionally, international collaborative efforts would help promote sustainability driven LIB processes. With advancement of artificial intelligence (AI), spent LIB recycling and a mindful use of LIBs considering their entire

life cycle can be done in a highly efficient way. Intelligent battery management system can aid in life cycle analysis of LIBs and can in turn improve the recycling process.

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