# A Perspective on the Sustainability of Cathode Materials used in Lithium-ion Batteries

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# Abstract

Electric vehicles powered by lithium-ion batteries are viewed as a vital green technology required to meet $CO_2$ emission targets as part of a global effort to tackle climate change. Positive electrode (cathode) materials within such batteries are rich in critical metals – particularly lithium, cobalt, and nickel. The large-scale mining of such metals, to meet increasing battery demands, poses concerns surrounding material exhaustion in addition to further environmental, social and governance (ESG) issues. In particular, unethical mining practices and political instability within the Democratic Republic of the Congo (the world's largest cobalt producer) have prompted research into cobalt-low and cobalt-free alternatives. This review aims to provide a holistic view of lithium-ion cathode development and inform advancements by highlighting the interdependencies across mining, material development, and end-of-life management. Whilst material sustainability is reported through supply and demand projections, the potential socio-environmental impacts of lithium-ion technology represent a hugely under-researched area amongst the aforementioned themes. Notably, the lack of attention paid towards future implications of increased nickel use across material management and development disciplines is also discussed.

# Introduction

High energy density lithium-ion batteries (LIBs) facilitate portable behaviours in modern society, contrived by a high-speed culture, that require us to communicate, work, and even charge 'on the go'. Beyond convenience, such technologies are taking centre stage in the environmental revolution through the ever-growing adoption of electrified modes of transport, as transportation currently accounts for 23% of global energy-related $CO_2$ emissions.[1] Electric vehicles (EVs) thus represent a rapidly expanding market, with at least 20% of road vehicles estimated to be electrically powered by 2030.[1] LIB technology takes great prominence within the automobile industry, due to its unbeatable electrochemical performance and lightweight, portable nature. Its impressive performance can be attributed, in part, to the low weight and small ionic radius of the $Li^+$ ions (0.76 Å), allowing fast ion transport. This fast transport, along with its low reduction potential (-3.04 V vs. standard hydrogen electrode (SHE)),[2] allows for high power density as well as volumetric and gravimetric capacity. Such properties are of critical importance for EVs.[3] With the increased demand for high energy density LIBs for EVs, comes reductions in battery cost and subsequent volatility in material supply. In light of the immense scale of transport electrification that is being proposed in order to meet $CO_2$ emission targets, considerable attention is being directed towards the socio-environmental and economic impact of such an increase in material demand. Of particular focus are lithium-ion cathode materials, many of which are comprised of lithium (Li), nickel (Ni), manganese (Mn), and cobalt (Co), in varying concentrations (**Fig. 1a**). The cathode constitutes more than 20% of LIBs overall cost and is a key factor in determining the energy and power density of the battery (**Fig. 1b**).[3,4] It is, therefore, vital to maximising the cathode performance whilst minimising its cost, to make EVs more accessible for society.

The high cost of cathode materials is largely attributed to the presence of cobalt - a rare and expensive element mined primarily in the Democratic Republic of the Congo (DRC), which has been deemed necessary in the past to deliver high energy densities in LIBs. For example, the active material within the commercial NMC111 cathode ($LiNi_{0.33}Mn_{0.33}Co_{0.33}O_2$) costs ca. £17 $kg^{-1}$, producing 3.88 kWh $kg^{-1}$.[5] This high cost is largely attributed to the relatively large amount of cobalt within the electrode (£ 25 $kg^{-1}$).[6] It is over 350 times greater than the cost of iron (£0.068 $kg^{-1}$),[7] which reflects its relative natural abundance. A combination of political instability within the DRC, social impacts within the mining sector, and supply chain volatility and ambiguity have driven a decrease in cobalt content in NMC cathodes (e.g. from NMC 111 to NMC811 ($LiNi_{0.8}Mn_{0.1}Co_{0.1}O_2$)) and potential future cathode materials ($LiNi_{0.5}Mn_{1.5}O_4$ spinels, $LiMO_2$ disordered rock-salts and $LiNi_{1-x}M_xO_2$ layered nickel-rich oxides).[8] Such a drastic shift to nickel-rich alternatives begs the question: 'In what way will decreasing cobalt and increasing nickel demand affect future supply amongst other environmental effects?'. Although this question remains largely undiscussed throughout the literature, the precarious environmental state and dire acceleration of EV consumption highlight the need for battery developers to place their research into a wider context to better inform material progression. With this in mind, this review aims to provide a more holistic insight into cobalt-low and cobalt-free cathode materials, thus considering material supply and demand amongst other environmental, social and governance (ESG) issues to provide a perspective on the future cathodes under development.

## Current Cathode technology and Material Development

The high transition metal content required to induce redox reactions to store charge in LIB cathodes leaves their formulations open to scrutiny, where the literature often highlights concerns surrounding lithium and cobalt supply risk. Research to overcome the main challenges faced by LIBs is underway with the exploration of alternative monovalent battery technologies such as sodium-ion[9] and divalent batteries, e.g. magnesium[10] and calcium[11] batteries. Yet, LIB technology will remain the market leader for the foreseeable future until such alternatives can offer parity in performance. Although the removal of lithium from cathode materials is unfeasible for present implementation, materials that require less lithium per kWh are preferable.

The start of the EV influx from 2015 saw that much of the LIB market was dominated by cathodes with high cobalt content, such as NMC111.[12] However, increased consciousness towards cobalt supply risk within the field of LIB development has resulted in the adaption of cathodes with reduced cobalt content, such as NMC811. Beyond reducing cobalt content, much research is invested into cobalt-free alternatives. Commercialised options available include lithium iron phosphate ($LiFePO_4$)[13] and lithium manganese oxide ($LiMn_2O_4$)[14], the use of which is often limited to certain applications due to unsatisfactory electrochemical performance for use in EVs (i.e. low energy-density and power-density, and poor cycle life in the case of LMO). This prompts research into further improving such cathodes for EV applications in addition to developing other potential future cathode materials. The aim for future LIB cathodes is, therefore, to minimise cobalt and lithium required whilst still maintaining, or better yet improving, electrochemical performance including energy density, power density, and long-term cycling stability. Such material development will be briefly outlined below.

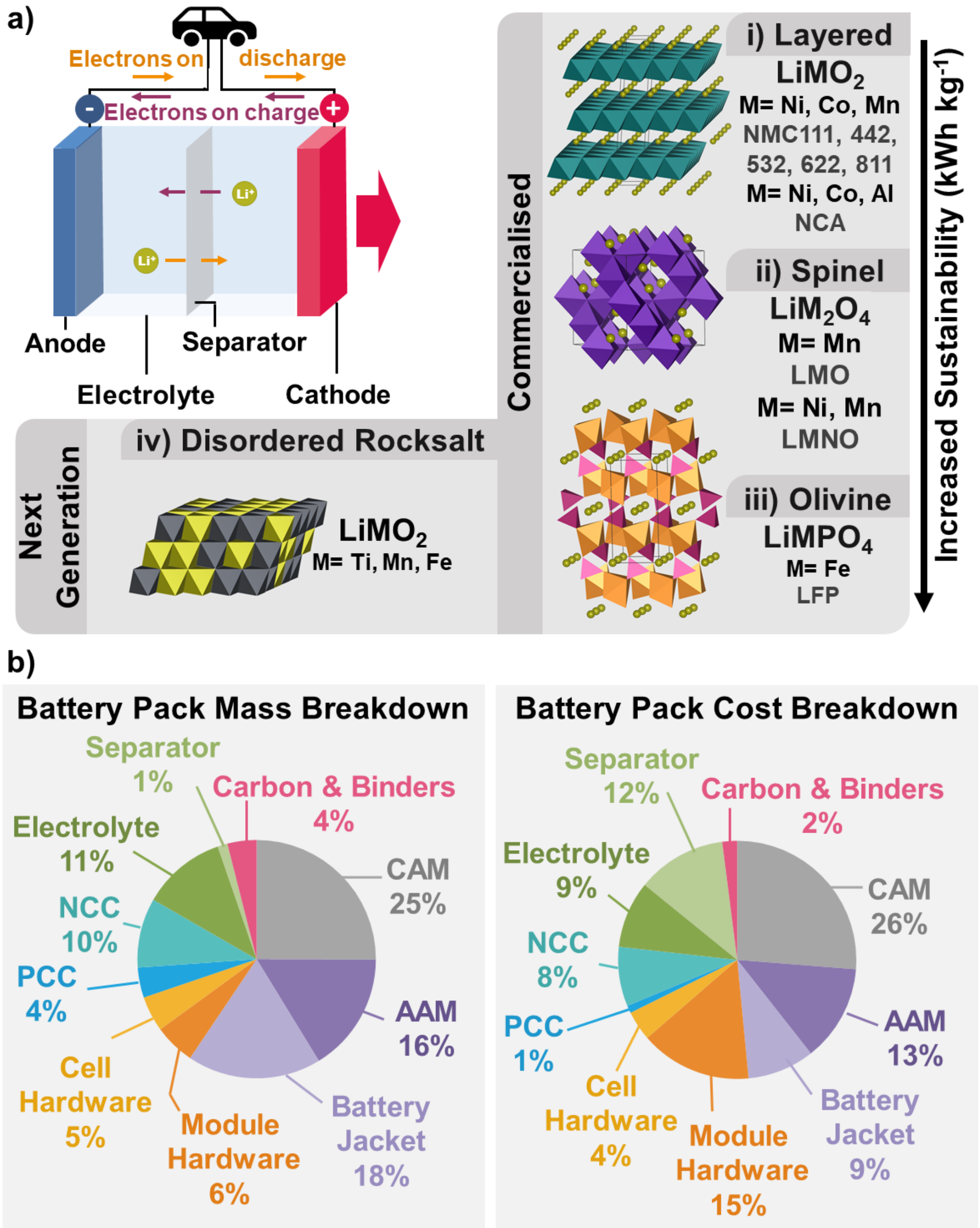


Figure 1. a) Schematic of a lithium-ion battery, showing the movement of electrons and lithium ions (green) during charge (purple) and discharge (orange) processes. Crystal structures of various cathode chemistries are indicated: i) layered structure, where teal octahedra represent M (M=Ni, Mn, Co for NMC and M=Ni, Co, Al for NCA), ii) spinel structure, where purple octahedra represent M (M=Mn for LMO and M=Ni, Mn for LNMO), iii) olivine structure, in which M=Fe for LFP, and iv) disordered rock salt structure, where cation mixing of M($d^0$) (grey) and Li (green) between the layers is observed (M= Fe, Mn, Ti). b) Typical mass and cost breakdown of an NMC battery pack, where CAM = cathode active material, AAM = anode active material, NCC = negative current collector and PCC = positive current collector. Charts produced with data from reference 15.

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## *Layered cathode materials*

Layered cathodes (**Fig. 1a**) represent the most widely researched cathode type for LIBs, where NMC-type cathodes ($LiNi_{1-x-y}Co_xMn_yO_2$) show particular prominence. The combination of nickel, manganese, and cobalt provides high specific capacity, low internal resistance, and high stability, respectively.[12] Although NMC111 is the most common, NMC-type cathodes with reduced cobalt content are gaining in importance to mitigate sustainability and cost implications associated with the critical element supply risk (see **Mining and Material Management** section). Progression through low-cobalt NMC cathodes has seen a variety of formulations including NMC442, NMC523, NMC622 and NMC811, which result in lowered pristine material costs (**Fig. 2a**), where in the case of NMC811, raw materials make up approximately 30% of production costs (**Fig. 2c**). In addition to the benefits related to decreasing cobalt concentration, the increased nickel concentration enhances capacity, with NMC811 showing an improved specific capacity of 200 mAh $g^{-1}$ when compared to NMC111 (160 mAh $g^{-1}$, both 4.3V vs. $Li/Li^+$).[12] Increasing the nickel content in these NMC-type cathodes, however, increases the reactivity of the cathodes due to the instability of nickel ions towards the liquid organic electrolyte and any trace moisture.[12] This prompts the need for additional cathode components to prevent degradation, such as electrode coatings, for example. Beyond simple surface coatings are advanced particle design strategies such as core-shell[16,17] and concentration gradient particles[17,18], in which nickel-rich NMC occupies the particle core to provide desirable electrochemical performance, whilst less reactive manganese-rich NMC dominates the particle surface (shell), providing enhanced stability against the electrolyte.[12] Commercially, NMC-type cathodes are often synthesised through a two-step co-precipitation reaction in which the metal hydroxide or carbonate is precipitated before sintering with stoichiometric amounts of lithium source (lithium carbonate or lithium hydroxide).[19] Whilst material costs may decrease due to reducing cobalt concentration, the manufacturing cost may increase due to the greater processing cost related to nickel and the use of, more expensive, lithium hydroxide as the lithium source required for the synthesis of nickel-rich cathodes (\$9.50 $kg^{-1}$ LiOH compared to \$7.75 $kg^{-1}$ $Li_2CO_3$, 2021).[20,21] NMC 712 shows an optimal elemental composition when considering a variety of factors including cost and abundance.[22] Considerations towards the increased $SO_x$ emissions associated with nickel increase must also not be overlooked (see **ESG Impacts section**).[23] Furthermore, the thermal safety of the NMC cathode with higher nickel contents, such as NMC811, is more hazardous due to the earlier exothermic onset temperature and largest exothermic heat generated.[24]

NCA cathodes ($LiNi_{1-x-y}Co_xAl_yO_2$) join NMC-type cathodes as front runners within the automobile industry. The NCA formulation has been optimised to 5 wt% aluminium (NCA-80,

$LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$), showing a comparable specific capacity to NMC811 (200 mAh $g^{-1}$, 4.3 V vs. $Li^+$/Li).[20] The lack of manganese in NCA materials (i.e. NCA-80,81 and 82) results in desirable capacity retention when compared to NMC811 as manganese ion dissolution is eliminated, whilst the incorporation of aluminium ions provides enhanced thermal stability.[20] Correspondingly, NCA is often the choice for 'long-range' EVs provided by Tesla, which boast ranges > 500 km.[12]

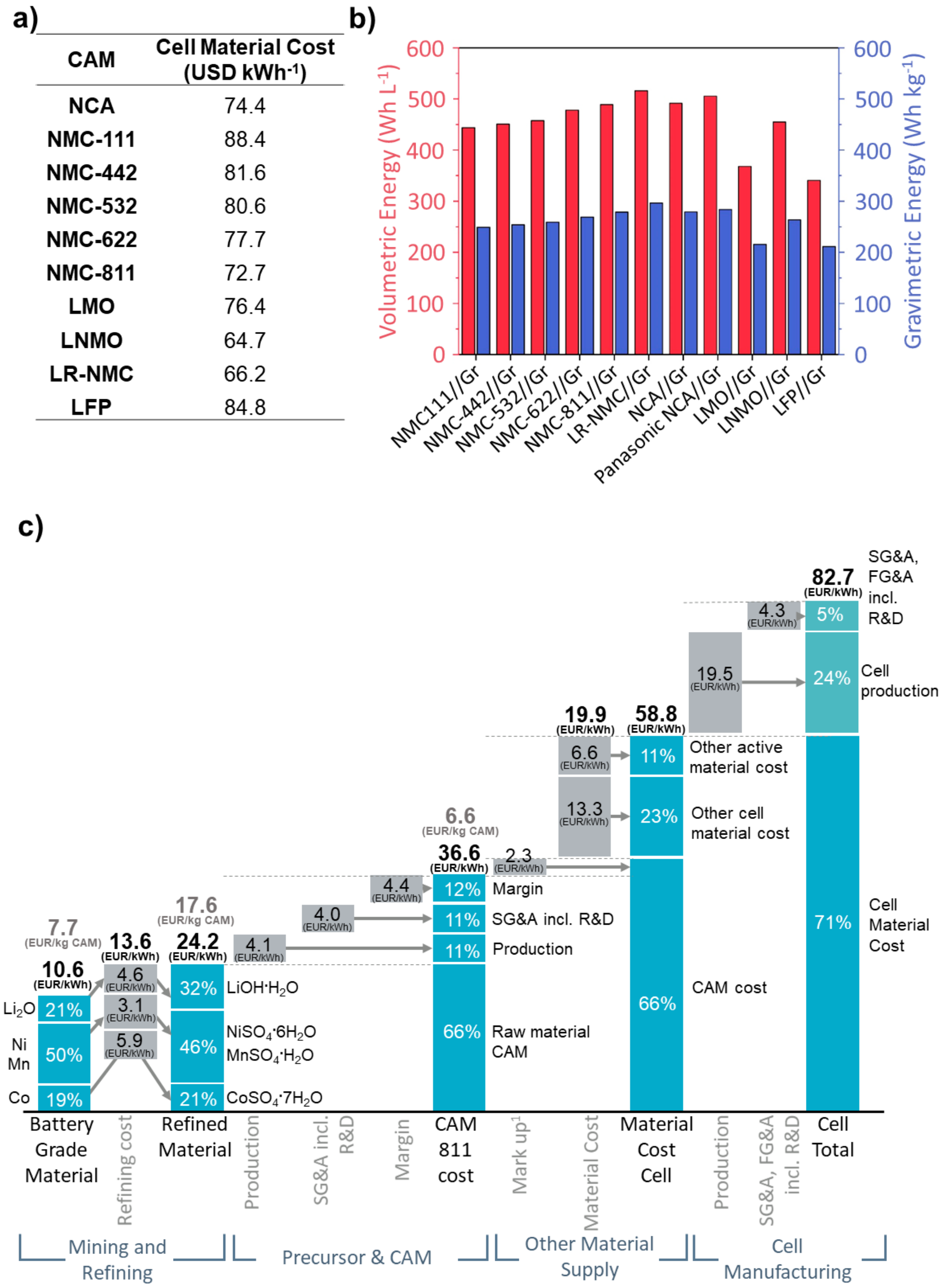



Figure 2. a) Estimated cell material cost based on production capacity of 1 GWh, data reproduced from 25, b) selected electrochemical performance parameters (volumetric and

gravimetric energy) from full cells with graphite (Gr) as anode and a variety of lithium-ion cathodes such as NMC111, 442, 532, 622 and 811, LR-NMC (lithium-rich NMC), NCA, LMO, LNMO and LFP. Reproduced with permission from 25, and c) Cost breakdown of an NMC811 prismatic cell produce in China considering costs related to mining and refining, production of Cathode Active Material (CAM), production of other cell components and cell manufacturing (SG&A = selling and general & administrative expenses, FG&A = factors that account for general & administrative expense, $Li_2O$ = lithium spodumene concentrate 6%, [1] = mark up of ca. 6.3% to account for efficiency losses between theoretical vs. nominal voltage.) Adapted with permission from 26.

Li-rich (LR) NMC type cathodes ($Li(Li_wNi_xCo_yMn_z)O_2$) exploit both cationic ($Ni^{2+/4+}$, $Co^{3+/4+}$) and anionic ($2O^{2-}/O_2^{n-}$ [n < 4]) redox activity allowing further improvements in capacity when compared to conventional NMCs (> 270 mAh $g^{-1}$).[19,27] Such a significant increase in capacity results in lower cell material cost (Fig. 2a).[25] These materials, however, suffer from capacity and voltage fade as well as voltage hysteresis and slow kinetics that result from the anionic redox. LR-NMC's with higher Ni content (i.e. LR-NMC811) are more effective at mitigating such issues.[27] More recently, disordered rock-salt (DRX) $LiMO_2$ cathodes (**Fig. 1a**) offer cobalt-free layered cathode that requires $d^0$ metal species and excess lithium.[29] These are, however, at a very early stage of research development. For sustainability reasons, iron,[30,31] manganese[32] and titanium[30,31,33] – based oxides are of particular interest. Substitution of oxygen by fluoride anions has shown to allow high reversible capacities (> 300 mAh $g^{-1}$) and energy densities (ca. 1000 Wh $kg^{-1}$, 1.5-5.0 V vs. $Li^+/Li$)[29] by averting the occurrence of irreversible oxygen redox reactions and/or $O_2$ loss.

## *Non-layered cathode materials*

$LiFePO_4$ (LFP), a cathode with an olivine structure (**Fig. 1a**), exhibits excellent cycle life and high thermal and electrochemical stability, due to the strong bond energy of the $PO_4$ tetrahedral units.[12] These properties, along with its inherently low cost and use of naturally abundant iron, make it an attractive cathode option for several battery applications. Its widespread adoption in EVs, however, is limited by its low energy density (120 Wh $kg^{-1}$) and poor electronic conductivity (ca. $10^{-9}$ S $cm^{-1}$), which despite low material cost results in relatively high cost per kWh.[20,25,34] LFP is typically synthesised through a two-step route in which the precursor is prepared through spray drying followed by calcination in an inert or mildly reducing atmosphere.[19] This is often coated with conductive carbon to improve the poor electronic conductivity.[35] The synthesis of nano-sized particles is also considered to improve electronic conductivity by decreasing the lithium-ion diffusion pathway.[36] Despite these drawbacks, LFP cathodes may still have a role in public transport, due to their high safety and fast charging times of ca. 2.5 h, and in less power demanding stationary storage.[12]

Spinel-type cathodes (**Fig. 1a**) provide an additional opportunity to eliminate cobalt, within certain battery applications, whilst also benefitting from decreased wt% of lithium when compared to layered transition metal oxides (e.g. NMC, NCA).[37] Their three-dimensional structure allows for facile lithium-ion diffusion and thus high-rate capability.[37] $LiMn_2O_4$ (LMO) represents the most widely researched spinel to have penetrated the EV market. The use of LMO is limited, however, by its low capacity and energy density, and short lifetime (due to structural instabilities upon cycling). Thus, LMO is often blended with NMC-type cathodes (for example, by automotive manufacturers Mitsubishi) to provide the high rate capability and low cost of LMO alongside the high capacity and improved cycling stability of NMC-type cathodes.[20] More recent research efforts have turned to focus on the high-voltage $LiNi_{0.5}Mn_{1.5}O_4$ spinel. The incorporation of nickel into the parent LMO spinel allows for high operating voltage and high energy density, through a two-electron $Ni^{2+/4+}$ redox couple (ca. 4.75 V vs. $Li^+/Li$).[37] This increase in energy density results in a decrease in cell material cost, despite the incorporation of a more expensive component (nickel), as less material is required per kWh.[25] As with LMO, however, LNMO is limited by structural instabilities on cycling in addition to incompatibility with commercial electrolytes resulting in electrolyte oxidation at such high voltage (> 4.5 V).[37] In order to compete with commercial cobalt-containing cathode materials, methods to improve such failure mechanisms are under investigation. These methods include various doping strategies[38,39], high-voltage electrolytes[40,41], surface coatings[42] and particle morphology optimisation[43]. Doping with abundant elements, such as iron at low concentrations, has not only shown to improve electrochemical performance, (particularly at high C rates) but could alleviate nickel demand which may prove beneficial when considering long-term supply vs. demand (see **Supply vs. Demand** section).[44]

# Mining and material management

## *Mining*

The mining of raw materials can have significant consequences for the resulting environmental, economic, and social impact of LIBs. Cathode materials constitute a considerable amount of the raw materials required for, and the cost of, LIBs. High cathode costs are a consequence of using critical elements such as lithium and cobalt. On the other hand, nickel and manganese are considered to be far less critical. Nevertheless, it is worthwhile considering supply, demand, and wider consequences of all constituent elements in cathodes to best project the outcome of rapid EV adoption.

Mining of lithium occurs primarily in South American countries, such as Chile and Argentina, in which lithium is extracted from brines and largely processed to form lithium carbonate ($Li_2CO_3$), which can then be converted into lithium hydroxide (LiOH). Brines containing lithium are estimated to represent 66% of global lithium resources (estimated to be 81 Mt by the U.S. Geological Survey, 2020).[45,46] Hard-rock extraction, on the other hand, from minerals such as spodumene, is largely employed in Australia. Whilst each of these countries focuses on only one extraction method, China uniquely produces lithium from both brine and hard-rock.[47] Unlike brines, spodumene can be directly transformed into LiOH, being approximately $ 500 $t^{-1}$ cheaper than LiOH from brine.[47] It is predicted that LiOH will constitute a large share of future demand due to its preferred use for long-range batteries.[47] The preferred use of LiOH over $Li_2CO_3$ is due to the instability of high nickel content NMC cathodes (NMC811) when synthesised with $Li_2CO_3$.[20] The use of LiOH in their synthesis, compared to $Li_2CO_3$, allows the use of lower synthetic temperatures, helping to maximise stability.[20] In addition to conventional sources such as hard-rock and brines, Tesla is hoping to extract lithium from clays using salt (sodium chloride). However, this source is often deemed unfeasible due to the low grade and high extraction cost.[48]

Cobalt mining is geographically concentrated in The DRC – home to the copper belt – where it is heavily mined, with China and Canada following as the 2nd and 3rd largest producers.[49] Cobalt is primarily produced as a co-product of copper mining (70% current supply, > 30% copper mine revenue) and a by-product of nickel mining (20% current supply, < 5% nickel mine revenue).[47,49] An estimated 15-20% of the DRCs cobalt supply is produced by small-scale artisanal miners who are not officially employed.[49] The role of the DRC as the main cobalt provider is predicted to remain stable, where they are projected to supply 62-70% from 2018 to 2030.[50] Future projections, however, suggest that cobalt supply as a by-product of nickel mining will increase. Shifting from co-product supply to by-product supply will ultimately reduce the interdependencies of cobalt on primary metal mining.[50] This, in turn, should improve the security of cobalt supply.

Nickel is mined, primarily in the Philippines, followed by Indonesia and Canada, as sulphide and laterite (oxide) ores.[23] Although laterites are more abundant, representing 70% of global stock, sulphides represent 60% of nickel supply due to the more complex, and thus more expensive, processing of laterites.[23] Unlike the aforementioned metals required for electromobility, manganese is plentiful, representing the third most abundant transition metal in the Earth's crust, of which 80% is mined in South Africa followed by Australia and China.[51]

The possibility of deep-sea mining is also being considered. However, widespread exploration of such mining is limited by the high upfront cost.[52] Furthermore, automotive companies such as BMW and Volvo have committed to avoid deep-sea mining due to the unclear effects on the fragile ocean eco-systems that are already under significant stress from overfishing, pollution and global warming.[52] Work across the social sciences is aiming to highlight and understand further issues surrounding social justice, vulnerability and ownership of deep-sea mining and mining areas.[53]

## *Supply vs. demand*

Various literature reports have attempted to predict supply vs. demand for metals used in cathode materials used in LIBs in order to elucidate potential future limitations. Such modelling and predictions prove difficult as the quantification of potential metal resources are highly dependent on public information provided by mining companies and other relevant sources, such as the U.S. and British geological surveys (USGS[54] and BGS[55]). Potential metal sources are often described in terms of resources and reserves. Resources represent a location in which a given metal is present in the Earth's crust. Reserves, on the other hand, represent resources that are economically feasible to mine.[56] Such feasibility is dependent on the deposit size, metal content and the extraction process required. For example, Bolivia contains the largest known lithium reserve (ca. 21 Mt[46]). However, lack of infrastructure for transportation and mining, limited quality of lithium-containing ore, and political barriers result in this area being under-mined.[57] Reserves are, therefore, dynamic – changing according to current socioeconomics, environmental policy and technology.[58] Estimations of supply are reliant on the number of deposits included from existing sources, sources that have announced future mining operations as well as projections towards potential unannounced mining operations (**Fig. 3**).[50] Demand modelling also depends on a large variety of factors including the time frame considered, the projected number of vehicles manufactured within this time frame, the share of different EV technologies in the market, the size of the EVs in question (kWh), the cathode material used and the weight of each metal per kWh (**Fig. 3**).[56] Any difference in the parameters chosen can result in significant modifications in the proposed supply vs. demand scenarios.

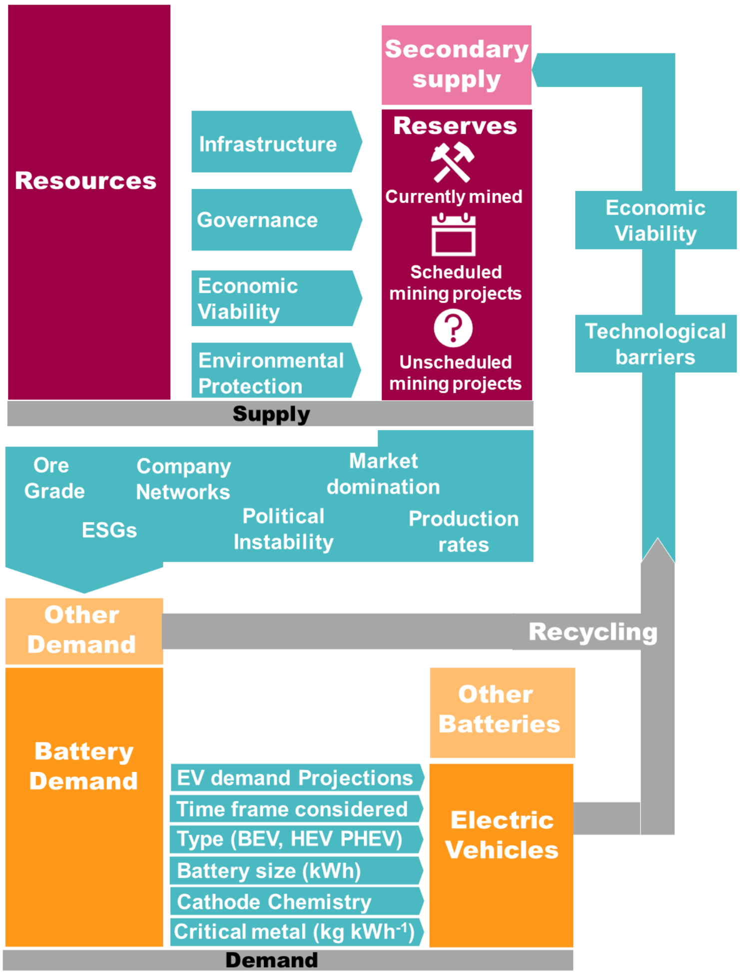



Figure 3. Schematic highlighting the considerations required for supply (purple, left) and demand (orange, right) modelling. Turquoise arrows indicate factors that influence the outcome of projections. Differences in factors chosen for modelling can result in significant differences in projected supply and demand.

Gruber et al. projected a total lithium demand over a period from 2010-2100 by modelling EV penetration, where annual EV growth beyond 2030 is anticipated to remain constant.[45] Such projections predict 100% EV penetration between 2083-2087. This results in an estimated lithium demand of 19.6 Mt, in which batteries dedicated for automotive applications account for approximately 65% of this demand. In this scenario, LIB recycling is estimated at 90%, with 90% recoverable lithium. Such recycling operations would significantly lower the strain on lithium mining. Evaluating lithium supply vs. demand, for 39 Mt of estimated in-situ lithium resource, suggest that supplies are sufficient to meet demand at least until 2100. This, however, is highly dependent on the success and implementation of lithium-ion recycling technology. Calisaya-Azpilcueta et al. took a different approach to model lithium supply chain through stochastic modelling, combining material flow analysis with both global sensitivity analysis and uncertainty analysis.[59] This allowed the identification of variables that had the most important effect on lithium distribution and EV production; lithium hydroxide production, from both lithium carbonate and hard rock, and traditional battery production. This work did not, however, consider stages beyond production. From their findings arose a probable scenario in which increasing demand is not covered by supply. For the time frame considered (2019-2025), this undersupply scenario was shown to be more likely to occur in 2025 than in 2021.[59] As a time frame beyond 2025 was not considered, that is not to say that lithium resources are predicted to be depleted by this time.

Fu et al. applied a series of scenario models for estimating supply and demand of cobalt over of short-term period (2015-2030). Their results indicated that, based on a high compound annual growth rate (CAGR), cobalt demand for EV LIBs accounts for 70% of battery demand by 2030 at 250 kt.[50] In addition to other battery applications and non-battery applications, in an aggressive high-demand scenario, it is projected that cobalt demand will reach 430 kt by 2030. This is closely matched to the projected 458 kt of supplied cobalt, under the same scenario conditions.[50] This work, therefore, envisages that cobalt supply will meet short-term demand. The possibility of recovering secondary cobalt through the recycling of electronics is estimated to provide an additional 17 kt into the supply chain (at a recovery efficiency of 100%).[50]

Elshkaki et al. postulate four different future scenarios and model the changes in nickel demand for each, where a collaborative ‘Equability world's scenario’ resulted in the highest demand (350% increase on 2010 by 2050) and lowest-demand in a 'security foremost’ scenario in which significant disparities exist (215% increase on 2010 by 2050).[23] In each of the four scenarios demand is expected to exceed reserves whilst remaining within the constraints of the estimated resources (150 Mt). This work predicts that nickel supplies will be sufficient to meet demand within the timeframe considered (2050). Concerns surrounding nickel for battery applications are often minor as battery demand represents only a small percentage of overall nickel demand when compared to lithium and cobalt required for battery applications (**Fig. 4**).[23] Nonetheless, reports have highlighted that although initial nickel supply may seem high, constraints defined by ore grade, governmental control and environmental and social pressures significantly limit the amount of nickel available for use in EV.[60] Their projections indicate limitations to nickel supply as early as 2027 when considering a low demand scenario. Conclusions and comparisons to literature reports for these scenarios, however, are not possible as the basis for such projections is not outlined. Despite this, it raises the importance of considering ore grade within supply and demand modelling as failure to do so may lead to misleading results.

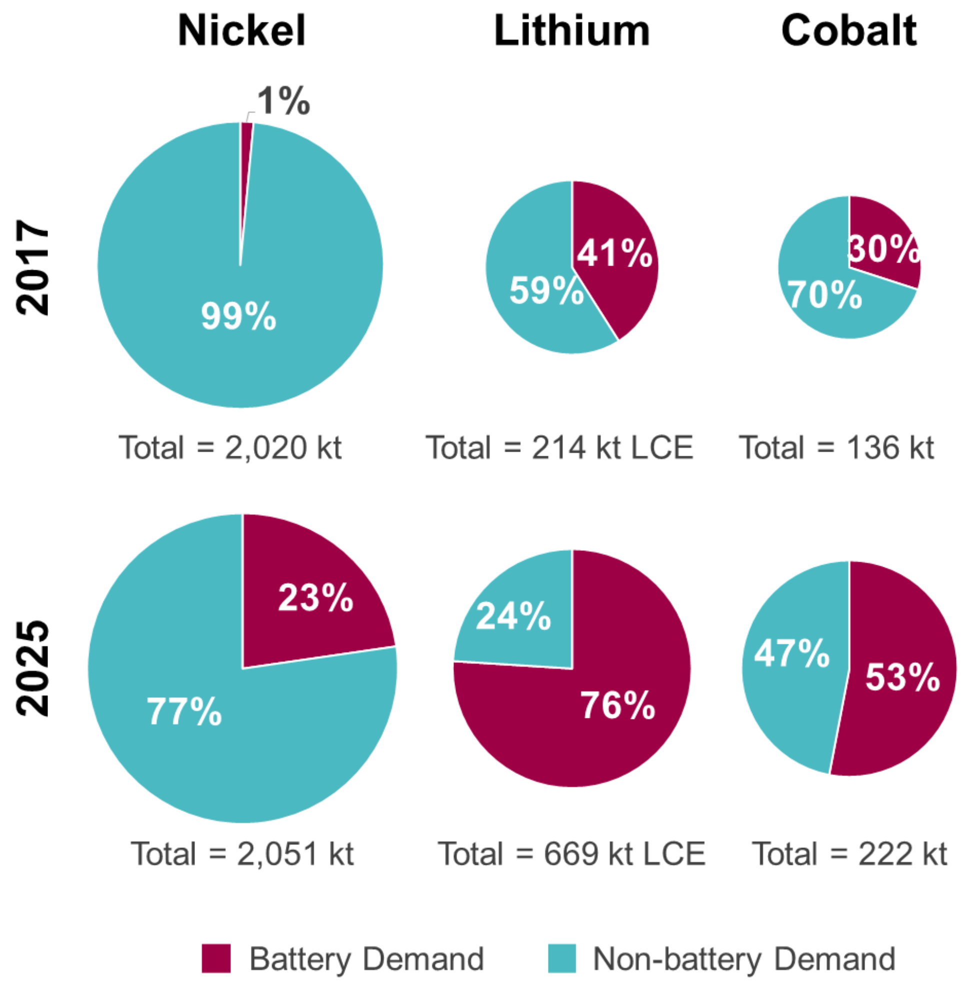



Figure 4. Predicted changes in battery and non-battery demand of nickel, lithium, and cobalt. Figures produced using data from references 12 and 24. LCE = lithium carbonate equivalents.

Being the third most abundant transition metal in the Earth's crust, supply vs. demand studies that focus on manganese alone are unsurprisingly difficult to come by. Unlike the literature above that discuss supply and demand of one focus element, work conducted by Habib et al. considered a range of materials required for EV production with a particular focus on cathode constituents.[61] This provides the benefit of comparing different elements under the same applied conditions. Three scenarios were modelled, based on representative concentration pathways (trajectories to predict climate futures, RCPs), which indicate the global warming delivered by a given concentration of $CO_2$ emissions (measured in W $m^{-2}$). Those considered are as follows: (1) 4.5 W $m^{-2}$ *(baseline*), where $CO_2$ emissions are required to start declining ca. 2045 and are expected to half between 2050 and 2100, (2) 2.6 W $m^{-2}$ (*stringent*), where

$CO_2$ emissions are required to start declining by 2020 and reach 0 by 2100, and (3) 3.4 W $m^{-2}$ (*moderate*), representing a scenario between 1 and 2.[62] Increased stringency to meet RCPs resulted in EVs constituting increased proportions of total 2050 passenger vehicles (23% of all vehicles electric in a baseline scenario, 32.6% in moderate and 73% in stringent). As expected, the increased in-use EV stocks significantly accelerate the reserve depletion of cobalt, lithium, and nickel, with cobalt reserves being depleted by 2035 under stringent modelling conditions. Other battery and EV constituents such as manganese, aluminium, iron, and copper, on the other hand, experience less significant depletion, retaining 90% (manganese, aluminium, iron) and 74% (copper) of original stocks up to 2050. As modelling followed an S-curve trend, all scenarios saw the highest demand for materials in 2035. A great disparity in material demand was seen between models, however, where cobalt demand was 11 times higher in the stringent scenario when compared with the baseline. This work identified nickel as well as lithium and cobalt as having high potential supply risk in the future.[61] No mention of ore grade is supplied within Habib et al.'s report, suggesting this supply risk is based on total nickel reserves as opposed to the 46% of nickel reserves that are acceptable for battery use.[60,61] With increasing nickel content in lithium-ion cathodes, greater attention must be paid to improving supply and demand modelling of not only lithium and cobalt but also, crucially, nickel.

Comparison between different models of supply vs. demand outlined above shows a large disparity in projected outcomes. Earlier attempts of modelling supply vs. demand lacked detail, often only considering one battery chemistry and EV type.[56] Recent developments show increased attention to specific EV and battery technologies employed, considering various cathode chemistries and relative EV battery sizes (kWh). Another area for uncertainty is non-battery applications. Whilst some works tried to also model non-battery applications, others do not, which would result in a gross underestimate of materials demand. The inclusion, however, adds further complexity and uncertainty to demand calculations. The sensitivity of modelling supply vs. demand renders outcomes doubtful, thus comparison studies, as conducted by Habib et al., may prove more beneficial.[61] Various time-frames used in reports make comparison difficult. As may be expected, with increased time, uncertainty increases due to the greater probability of significant changes in the supply and demand landscape.

Material demand is often modelled on different scenarios. These scenarios, however, are not consistent between reports. Whilst Speirs et al.[56] and Habib et al.[61] both considered scenarios based on $CO_2$ emission targets, the targets used were different with the former using IEA scenarios in which $CO_2$ emissions should see a 50% reduction by 2050 and the latter employing scenarios based on shared socio-economic pathways outlined by climate change researchers targeting different RCPs. This leads to significant differences in the anticipated EV and subsequent Li demand. Speirs et al. consider an EV market made up of BEVs and PHEVs, totalling 109 M vehicles in 2050. Varying material intensity within the EVs batteries resulted in a wide range of Li demand from 184-989 kt. Habib et al, on the other hand, predict EV demand between 2-3 M, with Li demand <100 kt for 2050. In addition to different scenarios used, different trends in EV adoption lead Habib et al. to predict a peak EV demand ca. 2030, whereas Speirs et al. observed a continual growth until 2050. Furthermore, Habib et al. included HEVs into their projections which use nickel-metal hydride batteries that are non-reliant on Li and so this will further reduce Li demand projections.

Focusing on projected cobalt demand for 2030, Habib et al., using RCP-based projections, predict EV demand from 30-70 million.[61] Fu et al. instead used compound annual growth rates (CAGR) of 5 and 10% when projecting EV demand, suggesting a range of approximately 10-21 million vehicles.[50] The former suggests a demand of approximately 500-5000 kt in 2030. The latter, on the other hand, predicts 235-430 kt of cobalt demand, where the higher limit is in line with the lower baseline limit projections of the former. Such a drastic increase in projected demand may be explained due to the far greater estimations of EV adoption to meet $CO_2$ targets where baseline efforts may be more probable unless significant policy is put in place. Despite the use of different scenarios, all recent works agree that supply will be sufficient for short-term to mid-term demand. A further drawback is that, although demand may appear to be within supply constraints, models often do not consider the rate of production for such critical metals. Lags in production rate may, therefore, pose a limiting supply factor.[63] It is evident from the works summarised, however, that cobalt poses the biggest depletion concerns followed by lithium.

Despite research efforts towards replacing LIBs with more sustainable alternatives (e.g. sodium-ion, magnesium, and calcium batteries), the requirement of LIBs for high energy density applications is likely to remain necessary for the foreseeable future as alternative technologies lag. This makes the complete removal of lithium unfeasible at present. Optimising formulations to minimise lithium content per kWh, however, can be investigated to minimise strain on lithium demand. Unlike lithium, cobalt can, and is, being substituted, largely by nickel and/or manganese (See **Material Development** section).[8] Despite no significant limitations predicted by literature reports on nickel supply in the near to mid-future, it would be worthwhile for modelling attempts to consider long-term supply vs. demand.

As seemingly abundant materials, iron (natural abundance (NA) = 56,300 ppm) titanium (NA = 5,650 ppm), and manganese (NA = 950 ppm) are viewed as worthwhile alternatives to nickel (NA = 190 ppm) and cobalt (NA = 25 ppm).[64] Studies that consider supply and demand for iron and manganese, focus on their use in steel and those that consider titanium, consider its use in pigments and within the aerospace industry.[65–67] From such studies it is difficult to extrapolate supply and demand to battery applications. As with nickel, however, battery applications form a small percentage of iron, titanium, and manganese demand. Supply and demand studies for iron focused entirely on supply and demand for steel, as it is estimated the 99% of the iron market lies within the steel industry.[66] As with nickel, widespread adoption of titanium- and iron- and manganese-based cathode materials will add further strain on to resources with already high demand. Here, we highlight the dangers of defining any given battery material as sustainable, as in doing so we lose foresight to future sustainability issues. It is clear from the extensive amount of resources required for successful EV penetration that a variety of cathode materials, used in conjunction throughout the industry, will be required to optimise sustainable development. More research into the potential impacts of increased iron, titanium and manganese battery demand should be considered pre-development, once again, to better inform materials development. Modelling approaches may be wise to consider a variety of up-and-coming materials (see **Material Development** section) to model the optimal share of each within the EV sector to best sustain resources. Such modelling attempts should allow anticipation of future bottlenecks. The undetermined electrochemical performance of novel materials when implemented in EV systems may, however, present some challenges and additional uncertainties.

## *Supply risk*

Supply risk is often assessed through product concentration, by-product dependency and political country risk (**Fig. 3**).[49] Whilst lithium and cobalt are both largely concentrated in South America and the DRC, respectively, companies located in China are largely responsible for the refinement of these raw materials for battery material production.[68,69] China has significantly increased investment into cobalt mining activities overseas in order to provide a domestic and steady downstream supply of raw materials. Chinese dominance of both raw and battery materials may lead to supply shortages if critical materials are leveraged in diplomatic disputes or reserved for their domestic use.[69] Therefore, country-level disruption to South American countries, the DRC or China could result in a significant impact on global lithium and cobalt supply resulting in high supply risk.[49] In addition to lithium and cobalt, environmental policies appearing throughout South East Asia banning raw ore exports or suspending nickel extraction in certain regions may pose a notable risk to nickel supply.[70] Increased insight into the environmental, social and governance (ESG) impacts of critical metal mining (see below) has led to increased consciousness towards responsible sourcing, which may further restrict resources available for use. Tesla has demonstrated the need for a secure supply chain by securing supply of both Ni and Li as these metals pose the greatest risk within their nickel-rich chemistries.[48,71]

Helbig et al. attempted to quantify the supply risk associated with a selection of metals used for battery applications.[72] From this study, it was determined that Li and Co posed the most significant supply risk (54% risk). Risk to Li supply was largely impacted by lack of sufficient recycling opportunities, whereas high Co risk was a result of political instability and by-product dependence. From the data presented, Ni (50%) and Mn (52%) show similar supply risk scores, with Ni risk largely dependent on supply reduction. Mn, on the other hand, shows a high score, despite its high natural abundance, due to its lack of substitutability. Ti was also evaluated, due to its use in lithium titanate anode materials ($Li_4Ti_5O_{12}$), and was shown to have a lower supply risk (43%). This work highlights the need to include Mn into supply considerations. Furthermore, understanding the impact of increased demand for Ti may be interesting for understanding the future implications of moving to Co- and Ni-free chemistries.

In order to reinforce supply chains, a more diverse stream of cobalt and lithium, in particular, will be necessary. Diversifying cobalt supply can be achieved through improved artisanal cobalt mining from >150 cobalt sites currently unmined, located in countries that do not presently mine.[49] Significant efforts should be made to improve the working conditions of artisanal mining through social and environmental sustainability measures as increased supply chain resilience could be achieved. The emergence of cobalt-primary mines, which has resulted from increased demand in the electronics sector, should help further improve cobalt security. Investment into lithium mining in Bolivia by foreign companies will considerably extend lithium reserves.[57]

In addition to geographical supply risk, company-based supply risk poses a potential threat. Companies that possess multiple links to other companies within the supply chain pose the biggest risk as a collapse in their supply could result in large-scale disruption. A large network of companies in the supply chain is, therefore, favourable to minimise such large-scale damage.[49] Any such shortages in supply may result in price increases. Cobalt shortages experienced between 2016 and 2017 saw cobalt prices approximately double (**Fig. 5a**). It is estimated that the cost of NCA and NMC increased by roughly 12.5%, as a result. A further decrease in cobalt content would limit the propagation of price and supply volatility to LIBs.[73] In contrast, nickel prices are far less volatile yet have seen a recent increase in prices, to their highest in six years due to increased demand for EVs (**Fig. 5a**). Lithium carbonate, on the other hand, experienced a drop in price between 2018 and late 2020 as increased production was not met by the required demand within EVs (**Fig. 5b**).[74] In order to sustain supply and demand, efforts must focus on developing electrode materials that are not reliant on scarce materials, extending battery lifetimes and improving reuse, repurpose, recycling and remanufacturing frameworks.[75] Recycling offers a reduced burden on mining by feeding into supply, reducing the primary metals required to meet demand. Supply risk also has the potential to benefit from recycling as secondary metal production can be exploited in countries without geological supply, thus diversifying the current supply chain. If, however, secondary supply is dominated by primary supplying countries, such as China, risks to supply would remain.[61] That being said, recycling will not alleviate strain within the near future given the lifetime of LIBs, rendering large material quantities in use until significant numbers of batteries reach end-of-life.[63]

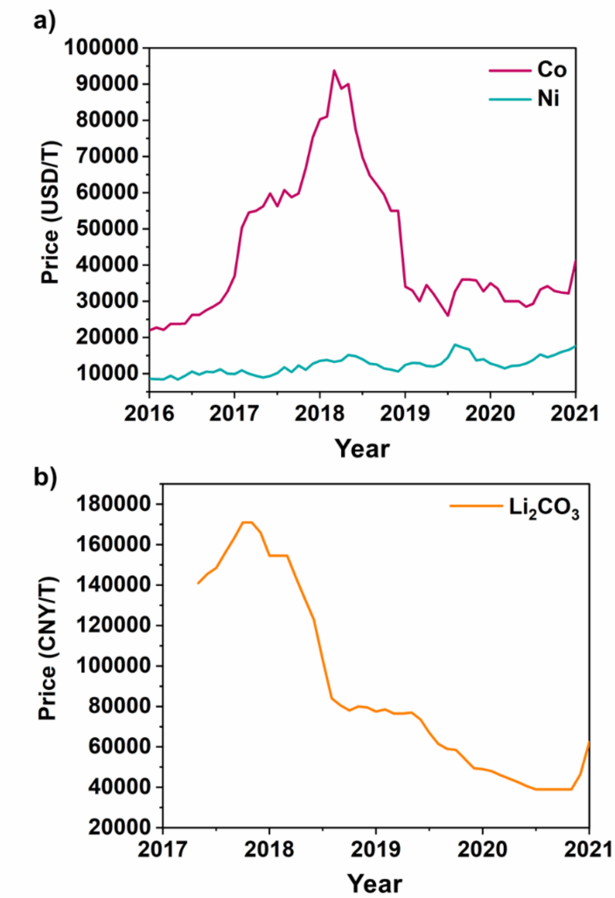
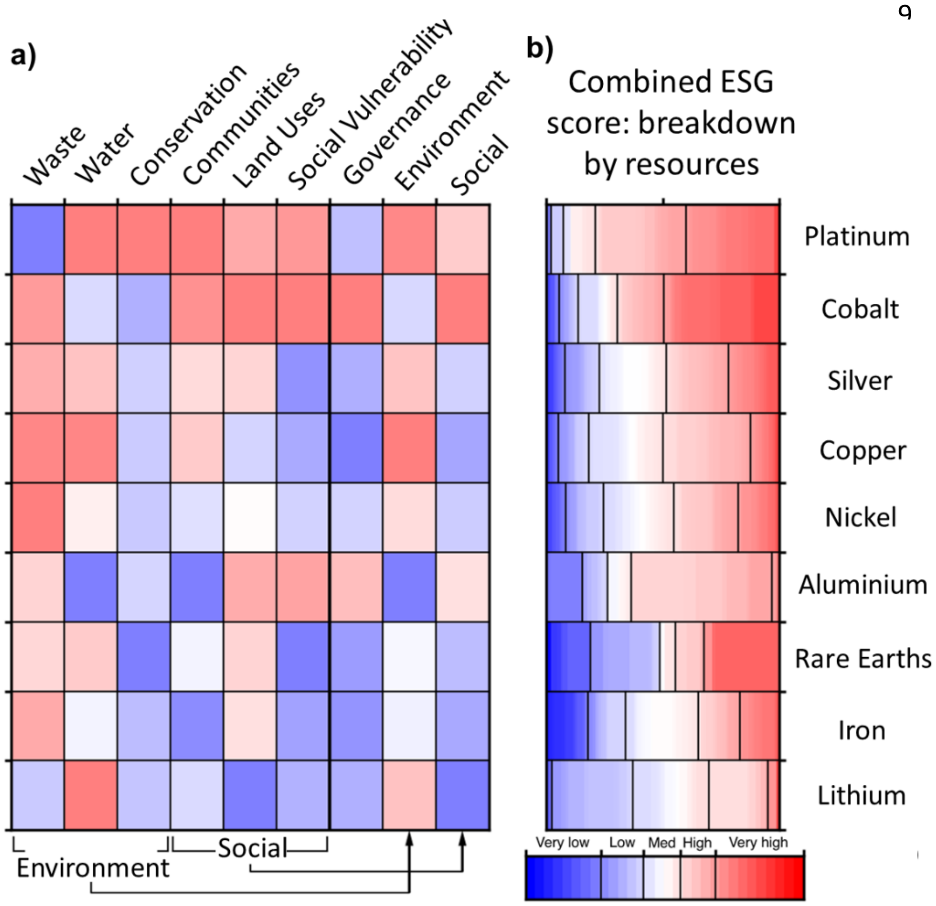



Figure 5. a) Price fluctuations of cobalt and nickel from 2016-2021 (USD/T). b) Price fluctuations of lithium carbonate traded in China, from 2017-2021 (CNY/T). Data from collected from references [76–78].

## *Environmental, social and governance (ESG) impacts*

Issues surrounding economics, supply, and demand appear to be the focus of LIB concerns, with a modest amount of literature reports on further environmental and societal matters. Sovacool et al. revealed the extreme risks to both environmental and public health, as well as social implications of gender discrimination and child labour in the DRC, exacerbated by the increasing adoption of 'green technologies' such as EVs.[79] Gender inequality in such areas, is allowed through mining hierarchies in which women appear very low, thus often carrying out the most strenuous yet poorly paid activities.[79] An estimated 23% of children within the DRC (many of whom are orphans) work within cobalt mining where they are exposed to physical, physiological and sexual abuse in order to provide for themselves and their families.[79] The long-lasting health impacts to societies within the vicinity of cobalt mines has been made apparent through the elevated cobalt levels in their blood and urine resulting in potential heart, lung, thyroid and blood complications.[73] Handling mining waste appropriately is also of utmost importance for ensuring the welfare of local residents. Waste from a previous mining plant after long storage was shown to have polluted surface and groundwater, atmospheric air, and soils with waste metals. The large number of heavy metals in water systems resulted in their presence within local foods.[80] In addition to toxicity concerns, tunnels dug for mining purposes lead to soil erosion and land instability.[79] Thies et al. considered child labour, corruption, occupational toxicity and hazards, and poverty as social risk factors throughout the supply chain of LIBs, through a social life cycle analysis – all of which were significantly reduced through the responsible sourcing of raw materials.[81] The supply chains investigated considered raw materials, cell components, battery cell production and battery pack production. Comparisons between supply chains that consisted of China- and Germany-based battery cell and battery pack production indicated greater risks in all the aforementioned areas from China-based battery production.[81] This highlights the value of responsibly sourcing and producing batteries and battery materials for the health and wellbeing of residents. Artisanal mining within the DRC has been highlighted in recent years due to child labour and unsafe mining conditions.[82] Banning of artisanal mining may, however, result in unintended harmful consequences as in poorer areas as it can be the only source of income for local residents.[82] In such communities more money can be earned through cobalt mining than through agriculture, creating a desire to mine with the hope of escaping poverty.[79] Although such issues accounted for a small amount of artisanal mining, further regulations and control are required to prevent such atrocities.[82]

Mining holds influence over ca. 50 million $km^2$ of terrestrial surface area, with 82% of mining area targeting critical materials required for clean energy production.[83] As the demand for different metals changes, the mining landscape will evolve with new mines opening, where desired ores are concentrated, and other mines closing due to declines in demand. The forced relocation of local residents to allow for mining expansion and the inhabitancy of old mining areas, due to decreased land quality, leaves a profound social and health impact on the local community.[84] In addition to residential areas, agricultural land and forestry also endures impairment due to mine expansion, with 14% of protected areas being within, or close to, metal mining areas.[84] The density of mining areas extracting critical metals that overlap with protected areas is far greater than the density of other mining areas which overlap. This indicates that with increased demand for critical metals for LIBs comes increased threats to biodiversity.[83] This provides additional ecological and environmental concerns beyond material exhaustion. Such socio-environmental considerations are often beyond the scope of the supply, demand, and economic concerns of mining activities. New and developing mining activities are thus encouraged to formulate considered mining plans that aim to assess nearby eco-systems, long-term effects and possible rehabilitation strategies.[84] It should be noted that issues will arise from the mining of most metals. It is, however, important to critically assess each in order to select those which pose minimal damage pre-use, during use and post-use. A comparative analysis into the ESG risk of a variety of transition metals used in green technologies, performed by Lèbre et. al, indicated that 70% of cobalt resources reside in areas with high ESG risks that are associated with a variety of factors across social vulnerability, land use, governance and waste.[85] Lithium, on the other hand, shows 65% of resources are in areas with low to medium ESG risk, where water use presents the biggest concern among the ESG factors (**Fig. 6**).[85] Nickel and iron show mining projects that are evenly divided across both high- and low-risk areas, where management and mitigation of ESG risks prove to be of critical importance for a global strategy to ensure minimal environmental and social impact with increasing demand.[85] Iron, when compared to nickel, cobalt and lithium shows very low

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ESG risk, with the biggest concerns stemming from toxic waste and land use (**Fig. 6**).[85] Primary environmental concerns related to cobalt, other than material exhaustion, are eutrophication and global warming potential, due to large amounts of electricity consumption for extraction. For nickel and manganese, on the other hand, greenhouse gas emissions (GHG) pose the biggest concern due to fossil fuel usage in mining, extraction and refining.[86] Access to sufficient renewable energy on the mining sites poses a hurdle for reducing GHG and global warming potential (GWP) as replacing existing supplies will prove time-consuming and costly.[86] Using high-grade ores can both economically and environmentally beneficial as processing requirements are lowered. As resources deplete, however, the extraction from low-grade ores will be inevitable.[86]

Nickel production, particularly from nickel sulphate ($NiSO_4$), is a very energy-intensive process that generates large amounts of sulphur dioxide ($SO_2$) during refinement.[87] This significantly increases the emissions related to LIB production. For this reason, the source of nickel production was shown to have a significant effect on the environmental impact through varying stringency on $SO_x$ capture, with Canadian refined nickel producing 0 kg $SO_x$ per kg $NiSO_4$ and Russian refined producing 2,902,991 kg $SO_x$ per kg $NiSO_4$.[87] This is dependent on the use of sufficient technology to capture and convert $SO_x$ emissions and highlights the importance of responsible sourcing. These figures are particularly alarming when considering that Russia produced 21.1% of battery-grade nickel in 2019, the largest producer of that year.[60] Life-cycle analysis conducted by Kallitsis et al. modelled three scenarios based on different NMC cathode chemistries (111, 622 and 811). Similar threats to humans and ecosystems are presented by novel chemistries.[88] Nickel sulphate production, however, resulted in an increase in all ecotoxicity categories considered as cathode nickel content increased. An overall decrease in the impact of LIBs using novel nickel-rich cathodes is provided through expected increased capacities. The prospect that the initial lifespan of novel positive electrodes may be inferior to existing ones should be considered and may limit the reduction of impact over the whole lifetime. Of the aspects considered, namely mining, extraction, processing, manufacture and assembly, battery production was found to have the most profound effect on environmental impacts. This is largely a consequence of non-renewable energy use in battery production.[88] The energy-intensive processing of nickel and cobalt ores accounts for a large proportion of energy consumption required to produce NMC-type cathodes. LMO and LFP cathodes, on the other hand, consume the most energy during the cathode preparation stage.[21]

With regards to the titanium dioxide ($TiO_2$) precursor, for which demand may increase if disordered rock-salt cathodes are to be successfully commercialised, production from starting materials such as rutile, ilmenite or titanium slag can be achieved through two methods: the chloride route and the sulphate route.[65] As with nickel processing, the use of sulfuric acid in the sulphate route poses potential environmental issues, which as previously mentioned can be eliminated through the use of sufficient mitigation practices.[89] Acid treatment, however, renders the sulphate route more costly.[89] The sulphate route is predicted to be the most common throughout Europe and China, whereas the chloride route dominates America.[65] The chloride route produces $TiO_2$ with higher purity and so such routes may be necessary to provide battery grade $TiO_2$.[65] Greater understanding into the impacts of Ti processing, and subsequent comparison with Ni, and Co, would be beneficial for understanding the true environmental gain of replacing Ni- and Co-rich chemistries.

# End-of-life and Waste Management

The possibility of a secondary metal supply from spent LIBs is commonly considered as a necessary addition to the extraction of raw materials in order to meet future demand. Waste LIBs from EVs, and other portable devices, are rapidly accumulating with little regulation in place to ensure safe and sufficient disposal within a coherent waste hierarchy scheme: prevention, reuse, repurpose, recycle, disposal.[90] Prevention, as previously discussed (see section **Current Cathode Technology and Material Development**), can be realised through material development, in which the amount of critical raw metal within cathode materials can be minimised. Subsequent improvement in performance of such materials is vital for lowering overall long-term demand.

Reuse involves the repair and/or remanufacture of spent LIBs for use in the same applications, whilst repurposed LIBs are to be used for less demanding energy storage (i.e. second-use).[90] For effective re-use, efficient battery management will be required in order to retrieve LIBs with approximately 80% state of health for subsequent repair and recirculation, likely as part of a battery leasing scheme.[90] LIB repair can involve identifying the cells within the battery pack (ca. 10%) with poorest state of health (SOH).[91] The identified cells can then be replaced, with fresh cells, avoiding replacement of the whole pack. Research into alternative charging methods illustrates a possibility of rejuvenating spent LIBs without disassembly, potentially reducing costs when compared to remanufacture and recycling waste streams. One such charging method is sinusoidal wave charging, as opposed to constant current charging, in which cycling to negative currents allows the reduction of solid electrolyte interface species at the anode surface, improving passivation.[92] This method has been shown to revive aged $LiFePO_4$ -based cells with SOH of 60-70%, 70-80%, and 80-90% by 18.7%, 9.5% and 4.2%, respectively. [92]

Whilst reuse would be intuitively favoured over repurposing due to less processing required, a study into the eco-efficiency of end-of-life (EOL) routes showed that repurposing allowed for greater reductions in cumulative energy demand, eco-toxicity, metal input and economic benefit.[91] This outcome largely came from the replacement of Pb-acid stationary storage with LIB stationary storage.[91] However, the quantity of LIBs that can be repurposed for second-use will far outweigh the second-use demand, due to the large amount of EVs going into circulation.[93] Furthermore, such repurposing will delay the retrieval of critical metals before the

LIBs are eventually discarded.[94] The successful implementation of battery recycling and critical metal recovery is, therefore, crucial for providing a sustainable supply of battery materials.

Each of the aforementioned EOL scenarios is limited by low collection rates (0-25% across different EU countries).[95] Such rates are proposed to be a result of insufficient EOL policy and public awareness of disposal protocols from which spent LIBs are often incorrectly disposed of or left as hibernating stock within society.[95] Under UK regulation the battery producer is responsible for paying for waste battery collection, treatment, recycling and disposal.[96] Whilst the disposal of LIBs into landfills is illegal under UK law[96], insufficient public awareness and lack of accessible disposal routes, such as kerbside collection, renders such practices inevitable.[97] The incorrect disposal of LIBs poses a significant safety concern due to the associated electrical, chemical and fire hazards that arise from damage to the battery packs and leaching of internal chemicals. Such events have seen approximately 48% of UK annual waste fires to be a result of waste LIBs.[97] This risk results in high transportation and processing costs such as manual disassembly, limiting the possibility for automated systems.[98] Whilst such manual disassembly may suffice in the short term, it will fail to cope with the greater influx of spent LIBs that is expected to come.[98] Automation within the disassembly line will, therefore, be paramount to the success of recycling operations.[98]

### *Recycling*

In addition to extending resources, the successful recycling of LIBs is suggested to alleviate other environmental concerns surrounding metal extraction, such as pollution, energy use and water use.[21,94,95,98–109] Beyond the environment, and as previously discussed (see section **Supply Risk**), a domestic secondary supply will reduce supply risk and mitigate price fluctuations, in addition to avoiding high transportation and processing costs of exporting and disposing of E-waste.[103] The price of recycled materials, however, may struggle to be competitive with primary resources, especially at the early stages of recycling development. This calls for incentives from policymakers to internalise social and environmental costs or subsidise recycled materials.[90] The benefits that recycling have on the social impacts of metal extraction, however, are largely unknown and may be a worthwhile investigation for future works to ensure the desired positive social impact.

Numerous reviews have been published within the last couple of years, in which various recycling methods under development are critically analysed.[21,94,95,98–109] Whilst an in-depth review of recycling methods is beyond the scope of the work presented herein, the technologies under development are briefly discussed and the trends in challenges identified and future outlooks proposed within a series of reviews are considered.

A variety of different recycling methods exist within the literature namely pyrometallurgy, hydrometallurgy, biometallurgy (**Fig. 7**) and direct recycling. Recycling first requires LIBs to be discharged, typically through the use of saturated sodium chloride solutions. If disassembling under inert conditions (e.g. under argon), however, such discharging is not necessary.[107] Mechanical separation is then used to dismantle the battery into its different components, from which the cathode material is extracted and further treated. Pyrometallurgy involves the heat treatment of recovered cathode materials to form an alloy of Cu, Co, Ni and Fe. Li and Al, on the other hand, are contained with the remaining slag from which they are difficult to extract. Due to the simplicity of pyrometallurgy, it is an attractive choice for recycling operations. However, the use of high temperatures and the release of significant greenhouse gasses limits its eco-friendliness. Hydrometallurgy involves the selective dissolution, leaching, separation and purification of metals from waste cathode materials. Typical leaching agents include $H_2SO_4$, $HNO_3$ and HCl.[107] Research into organic leaching agents, however, (such as oxalic and citric acid) is gaining importance in order to provide a more environmentally friendly alternative.[110] Bio-metallurgy uses microbiological processes that can produce organic or inorganic acids to extract critical metals.[99] Cathodes can then be resynthesized from the leachate solution via a co-precipitation or sol-gel method, which can simplify separation and purification steps.[110] Both hydrometallurgy and bio-metallurgy have the advantage of being able to recover Li, unlike pyrometallurgy. However, the high volumes of effluents produced require treatment before disposal.[107] Whilst bioleaching provides an eco-friendly and energy-efficient method, its poor adaptability and leaching conditions required currently limit its suitability for industrial applications.[110] Direct recycling poses a method in which the crystal structure can be retained thus improving the economic feasibility in addition to lowering environmental impacts.[111,112]

The cost of different recycling methods is comprised of labour costs, material costs and utilities amongst additional expenses such as tax, rent, insurance, and maintenance (**Fig. 7b**).[113] Leaching chemicals required for hydrometallurgical recycling result in higher material costs, whereas pyrometallurgical recycling is more labour intensive with higher utility costs, resulting in a higher overall cost in comparison. The financial viability of hydrometallurgical, pyrometallurgical and direct recycling methods are impacted by a series of factors including transport distances, labour cost, disassembly cost, recycling capacity and revenue generated from recovered materials. Although direct recycling is predicted to be slightly more expensive than hydrometallurgical recycling, higher net profit is anticipated with increased scaling due to increased revenue through higher material recovery. In comparison to European countries, such as Belgium and the UK, China and South Korea show lower recycling costs due to lower labour and general expenses costs. Despite this, an analysis into possible recycling routes for spent UK LIBs revealed that, due to high transportation costs, recycling abroad is uneconomic regardless of the cell chemistry and recycling method adopted.[113]

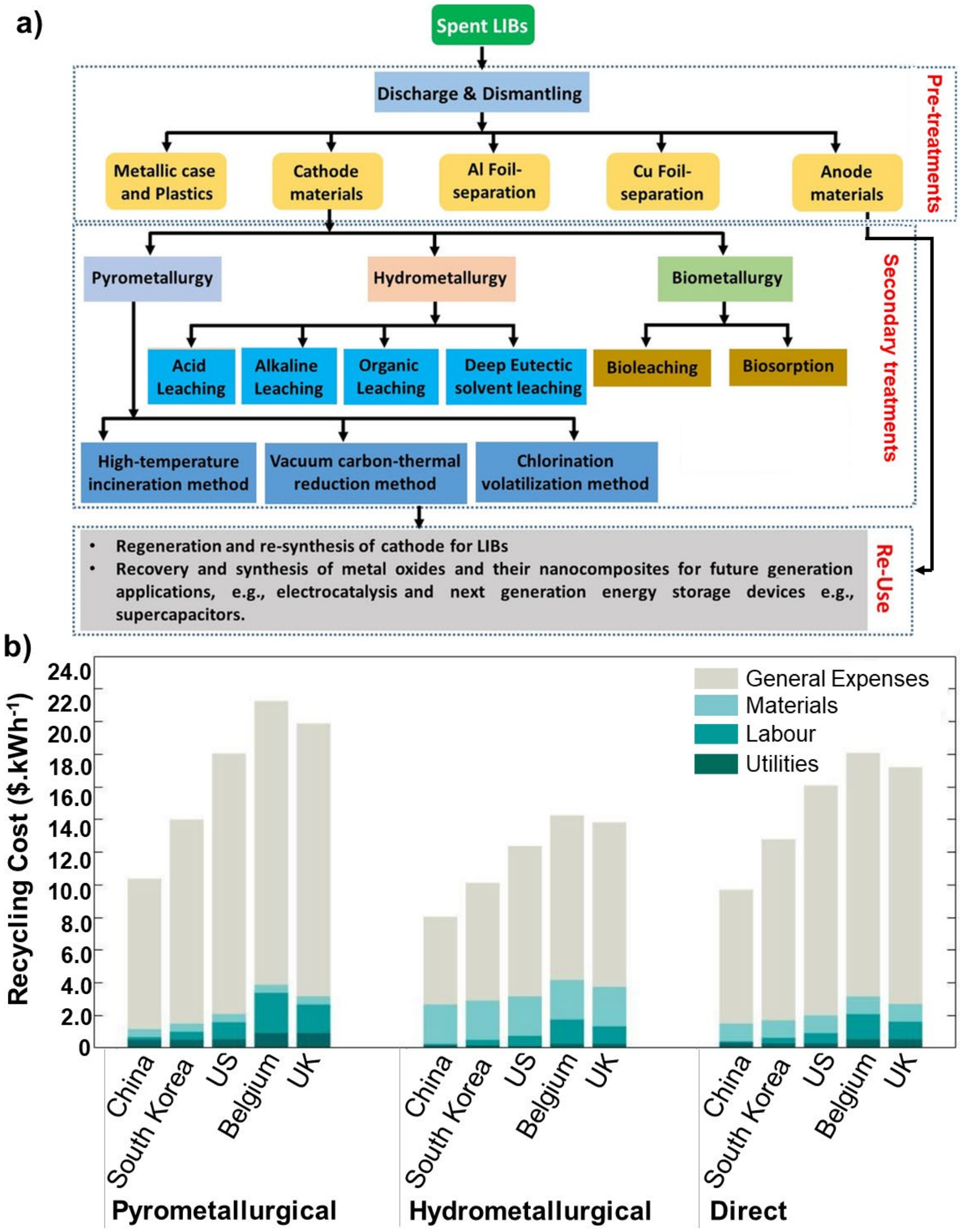


Figure 7. a) Schematic illustrating the closed-loop approach to LIB EOL through recycle and reuse. Reproduced with permission from 94 (copyright Wiley Materials), b) Breakdown of recycling cost for a 240 Wh $kg^{-1}$ NCA battery pack, reproduced with permission from 113.

The EOL of LIBs is confronted with many challenges spanning across social, environmental, economic, political, technical and chemical domains. Technical concerns dominate recent reviews, with barriers to automation seen as significant challenges.[21,94,95,98–109] Such barriers include the non-uniformity in cell designs adopted by different manufacturers. Furthermore, the large variety of cell chemistries used in LIBs require sorting before recycling can begin. Whilst a mixed market of battery materials may be beneficial for conserving resources, a wide variety of chemistries in circulation renders highly specific recycling techniques inadequate. Lack of labelling systems on battery packs makes pre-sorting challenging, and introduces additional safety concerns as LIBs can enter Pb-acid battery waste streams accidentally.[101] Of the reviews considered,[21,94,95,98–109] the technical barriers to widespread adoption of LIB recycling identified were ubiquitous, with each highlighting the need for; 1) sufficient labelling systems for easy identification, 2) standardisation of cell material, cell design and processing, and/or greater flexibility in the recycling processes, 3) minimisation of components, 4) screening, health monitoring and sorting methods and 5) automation in the disassembly line. It was acknowledged by the majority of reviews that many of these challenges require necessary intervention from policy-makers to provide a clear recycling industry chain and introduce sufficient regulations for the safe transport and handling of waste LIBs.[93,94,98–102,106,108]

Whilst recycling offers a potential secondary supply of materials, amongst other benefits, it is important to consider net changes in energy consumption when recycled materials are implemented. With a few exceptions,[99,108,114] environmental concerns related to recycling are largely underexamined and, if so, addressed qualitatively. Conclusions made by Huang et al. highlighted the need for further quantification of environmental damage/benefit including the quantification of waste and emissions.[108] For example, a study by Ciez et al. indicates pyrometallurgical and hydrometallurgical recycling processes do not pose significant environmental benefits when considering resulting reductions in greenhouse gas emissions.[73] For more environmentally friendly cathode technologies, such as $LiFePO_4$ (LFP), no amount of recovered LFP is sufficient to offset GHG emissions that result from both the recycling process and the incineration of other waste components. Furthermore, the decrease in cobalt concentration reduces the economic viability of such processes, perhaps limiting the recyclability of lithium and nickel. Having said that, if nickel resources begin to deplete to the levels that cobalt is currently experiencing, the economic viability of such recycling will inevitably increase.

As previously mentioned, direct recycling poses a method with greater economic and environmental benefits.[111,112] Recent works demonstrated the possibility of directly recycling LFP[112] and LMO[111] cathodes, in which life-cycle analysis showed a reduction in both GHG emissions (ca. 70%) and energy usage (> 75%). A critical review of recycling techniques, performed by Piątek et. al, revealed that, within the principles of green chemistry and circular economy, solutions presented are often very unsustainable.[99] Whilst recycling is key for materials sustainability, this adds another level of complexity to the holistic lithium-ion sustainability problem whereby recycling efforts must employ technologies that do not pose additional negative environmental and social issues.

# Conclusion and outlook

The issues presented by the widespread use of LIBs cover a wide range of sectors from onset through to end-of-life. Amongst them, mining and material management, socio-environmental life cycle analysis, material development and end-of-life management, outlined herein, are crucial for understanding and mitigating concerns surrounding supply risk and environmental, social and governance (ESG) issues. Whilst each one of these sectors plays an important role in LIB research, they are often considered as individual entities without additional thought to the other contributors. This review aims to place such material development into the wider context of ESG factors, in order to better inform cathode material development.

Progression towards ‘sustainable’ cathode materials within the industry has seen a shift to nickel-rich chemistries. However, the dependence of LIBs on high-grade nickel ore may pose a limit to supply. Supply and demand projections that consider ore grade will, therefore, be vital in assessing the Ni resources available for battery applications. The increasing complexity of the EV and wider battery market results in an increased number of parameters to be considered, in which ore grade, varying EV battery types and sizes used, and non-battery applications will demand greater attention. With an increased number of parameters, however, comes increased uncertainty in the results obtained. It is thus important to critically analyse previous models against real-time supply and demand data in order to determine their accuracy and provide an explanation for discrepancies to allow for the development of improved, and eventually standardised models. Standardisation of such modelling would prove beneficial for comparing between different elements and can be translated to other elements contained within newer cathode chemistries, to highlight changes in material sustainability with cathode composition. Due to the dynamic nature of available reserves, increased efforts to strengthen supply chains (e.g. establishment of more Co-primary mines), and the exploration of new mining opportunities (e.g. deep-sea and lithium clay mining), long-term models previously considered may require reassessment to account for such changes.

The~~is~~ potential limit to supply, in addition to geopolitical and company-based supply risk, may add further strain to cathode and LIB supply chains. Such supply may, however, benefit from the successful implementation of reuse, repurposing and recycling in order to extend the use of critical metals in stock and better distribute secondary resources that do not have such a significant dependence on geographical location. Implementation at a large scale, however, is limited by poor financial viability and lack of automation. Thus, developing simple and low-cost methods with increased recovery rates is vital for ensuring a secondary supply. Financial viability can be further improved by establishing domestic LIB waste schemes by avoiding high transportation costs, in which sufficient policy surrounding LIB waste management, increased recycling capacity and increased public awareness will be key. Whilst a secondary supply is crucial, the additional environmental impacts of recycling, such as waste and emissions, adds further complexity. Current literature lacks quantification of such impacts which is necessary for critically assessing and comparing various recycling methods.

Beyond material sustainability, further efforts are required to ensure environmental sustainability of Ni used in LIBs by introducing sufficient international regulation on sulphate capture to prevent additional damage caused by $NiSO_4$ processing emissions. It is therefore expected that in the future, more sustainable battery chemistries based on Co-free and low-Ni content materials focused on Fe, Mn and Ti elements will provide both socio-economic and environmental gain. However, a foreseeable practical research challenge will be engineering cathode materials with adequate elemental compositions that can achieve comparable or even better performance metrics than well-established and commercialised cathode materials. Similarly, these new materials will require a critical assessment on ESG issues to encourage sustainability progression and successful and responsible use in LIBs for EVs.

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# References

1. United Nations. Paris Declaration on Electro-Mobility and Climate Change & Call to Action. 2 (2015).

2. Vignarooban, K. *et al.* Current trends and future challenges of electrolytes for sodium-ion batteries. *Int. J. Hydrogen Energy* **41**, 2829–2846 (2016).

3. Nitta, N., Wu, F., Lee, J. T. & Yushin, G. Li-ion battery materials: Present and future. *Mater. Today* **18**, 252–264 (2015).

4. Desjardins, J. The Cathode is the Key to Advancing Lithium-ion Technology. *Visual Capitalist* https://www.visualcapitalist.com/cathode-advancing-lithium-ion/ (2017).

5. Ahmed, S., Nelson, P. A., Gallagher, K. G., Susarla, N. & Dees, D. W. Cost and energy demand of producing nickel manganese cobalt cathode material for lithium ion batteries. *J. Power Sources* **342**, 733–740 (2017).

6. Cobalt Price. *Metalary* https://www.metalary.com/cobalt-price (2020).

7. Metallary. Iron Price. https://www.metalary.com/iron-price/.

8. Gourley, S. W. D., Or, T. & Chen, Z. Breaking Free from Cobalt Reliance in Lithium-Ion Batteries. *iScience* **23**, 101505 (2020).

9. Kim, S. W., Seo, D. H., Ma, X., Ceder, G. & Kang, K. Electrode materials for rechargeable sodium-ion batteries: Potential alternatives to current lithium-ion batteries. *Adv. Energy Mater.* **2**, 710–721 (2012).

10. Mohtadi, R. & Mizuno, F. Magnesium batteries: Current state of the art, issues and future perspectives. *Beilstein J. Nanotechnol.* **5**, 1291–1311 (2014).

11. Gummow, R. J., Vamvounis, G., Kannan, M. B. & He, Y. Calcium-Ion Batteries: Current State-of-the-Art and Future Perspectives. *Adv. Mater.* **30**, 1–14 (2018).

12. Zeng, X. *et al.* Commercialization of Lithium Battery Technologies for Electric Vehicles. *Adv. Energy Mater.* **9**, 1–25 (2019).

13. Yamada, A., Chung, S. C. & Hinokuma, K. Optimized LiFePO4 for Lithium Battery Cathodes. *J. Electrochem. Soc.* **148**, A224 (2001).

14. Gummow, R. J., de Kock, A. & Thackeray, M. M. Improved capacity retention in rechargeable 4 V lithium/lithium-manganese oxide (spinel) cells. *Solid State Ionics* **69**, 59–67 (1994).

15. Nelson, P. A., Ahmed, S., Gallagher, K. G. & Dees, D. W. Modeling the Performance and Cost of Lithium-Ion Batteries for Electric-Drive Vehicles, Third Edition. *Argonne Natl. Lab.* 1–116 (2019).

16. Sun, Y. K., Myung, S. T., Park, B. C. & Amine, K. Synthesis of spherical nano- To microscale core-shell particles Li[(Ni 0.8Co 0.1Mn 0.1) 1-x(Ni 0.5Mn 0.5) x]O 2 and their applications to lithium batteries. *Chem. Mater.* **18**, 5159–5163 (2006).

17. Hou, P., Zhang, H., Zi, Z., Zhang, L. & Xu, X. Core-shell and concentration-gradient cathodes prepared via co-precipitation reaction for advanced lithium-ion batteries. *J. Mater. Chem. A* **5**, 4254–4279 (2017).

18. Yoon, S.-J., Park, K.-J., Lim, B.-B., Yoon, C. S. & Sun, Y.-K. Improved Performances of Li[Ni 0.65 Co 0.08 Mn 0.27 ]O 2 Cathode Material with Full Concentration Gradient for Li-Ion Batteries . *J. Electrochem. Soc.* **162**, A3059–A3063 (2015).

19. Schmuch, R., Wagner, R., Hörpel, G., Placke, T. & Winter, M. Performance and cost of materials for lithium-based rechargeable automotive batteries. *Nat. Energy* **3**, 267–278 (2018).

20. Li, W., Erickson, E. M. & Manthiram, A. High-nickel layered oxide cathodes for lithium-based automotive batteries. *Nat. Energy* **5**, 26–34 (2020).

21. Dunn, J. B. *et al.* Material and Energy Flows in the Production of Cathode and Anode Materials for Lithium Ion Batteries. *Argonne Natl. Laborartory* **53**, 1689–1699 (2015).

22. Shang, K. Nickel sulphate: In high-nickel batteries, safety comes into question. *Roskill* https://roskill.com/news/nickel-sulphate-in-high-nickel-batteries-safety-comes-into-question/ (2020).

23. Elshkaki, A., Reck, B. K. & Graedel, T. E. Anthropogenic nickel supply, demand, and associated energy and water use. *Resour. Conserv. Recycl.* **125**, 300–307 (2017).

24. Gong, J., Wang, Q. & Sun, J. Thermal analysis of nickel cobalt lithium manganese with varying nickel content used for lithium ion batteries. *Thermochim. Acta* **655**, 176–180 (2017).

25. Wentker, M., Greenwood, M. & Leker, J. A bottom-up approach to lithium-ion battery cost modeling with a focus on cathode active materials. *Energies* **12**, 1–18 (2019).

26. Bernhart, W. Battery Recycling is a Key Market of the Future: Ist it also an Oppourtunity for Europe? *Roland Berger* https://www.rolandberger.com/en/Insights/Publications/Battery-recycling-is-a-key-market-of-the-future-Is-it-also-an-opportunity-for.html (2019).

27. Ramesha, R. N., Bosubabu, D., Karthick Babu, M. G. & Ramesha, K. Tuning of Ni, Mn, and Co (NMC) content in 0.4(LiNixMnyCozo2)·0.4(Li2MnO3) toward stable high-capacity lithium-rich cathode materials. *ACS Appl. Energy Mater.* **3**, 10872–10881 (2020).

28. Li, W., Lee, S. & Manthiram, A. High-Nickel NMA: A Cobalt-Free Alternative to NMC

and NCA Cathodes for Lithium-Ion Batteries. *Adv. Mater.* **32**, 1–6 (2020).

29. Clément, R. J., Lun, Z. & Ceder, G. Cation-disordered rocksalt transition metal oxides and oxyfluorides for high energy lithium-ion cathodes. *Energy Environ. Sci.* **13**, 345–373 (2020).

30. Yang, M. *et al.* Nanostructured cation disordered Li2FeTiO4/graphene composite as high capacity cathode for lithium-ion batteries. *Mater. Technol.* **31**, 537–543 (2016).

31. Chen, R. *et al.* Identifying the redox activity of cation-disordered Li-Fe-V-Ti oxide cathodes for Li-ion batteries. *Phys. Chem. Chem. Phys.* **18**, 7695–7701 (2016).

32. Freire, M. *et al.* Investigation of the exceptional charge performance of the 0.93Li4-: XMn2O5-0.07Li2O composite cathode for Li-ion batteries. *J. Mater. Chem. A* **6**, 5156–5165 (2018).

33. KITAJOU, A. *et al.* Improvement of Cathode Properties by Lithium Excess in Disordered Rocksalt $Li_{2+2x}Mn_{1\−x}Ti_{1\−x}O_4$. *Electrochemistry* **84**, 597–600 (2016).

34. Myung, S. T. *et al.* Nickel-Rich Layered Cathode Materials for Automotive Lithium-Ion Batteries: Achievements and Perspectives. *ACS Energy Lett.* **2**, 196–223 (2017).

35. Ding, Y., Cano, Z. P., Yu, A., Lu, J. & Chen, Z. Automotive Li-Ion Batteries: Current Status and Future Perspectives. *Electrochem. Energy Rev.* **2**, 1–28 (2019).

36. Lei, Z. *et al.* High performance nano-sized LiMn1-:XFexPO4 cathode materials for advanced lithium-ion batteries. *RSC Adv.* **7**, 43708–43715 (2017).

37. Liang, G., Peterson, V. K., See, K. W., Guo, Z. & Pang, W. K. Developing high-voltage spinel LiNi 0.5 Mn 1.5 O 4 cathodes for high-energy-density lithium-ion batteries: current achievements and future prospects . *J. Mater. Chem. A* (2020) doi:10.1039/d0ta02812f.

38. Shin, D. W., Bridges, C. A., Huq, A., Paranthaman, M. P. & Manthiram, A. Role of Cation Ordering and Surface Segregation in High-Voltage Spinel LiMn LiMn 1.5Ni 0.5-xM xO 4(M = Cr, Fe, and Ga) Cathodes for Lithium-Ion Batteries. *Chem. Mater.* **24**, 3720–3731 (2012).

39. Fey, G. T. K., Lu, C. Z. & Prem Kumar, T. Preparation and electrochemical properties of high-voltage cathode materials, LiMyNi0.5-yMn1.5O4 ( M = Fe, Cu, Al, Mg; y = 0.0-0.4). *J. Power Sources* **115**, 332–345 (2003).

40. Zheng, Q. *et al.* A cyclic phosphate-based battery electrolyte for high voltage and safe operation. *Nat. Energy* **5**, 291–298 (2020).

41. Zou, Z., Xu, H., Zhang, H., Tang, Y. & Cui, G. Electrolyte Therapy for Improving the Performance of LiNi0.5Mn1.5O4 Cathodes Assembled Lithium-Ion Batteries. *ACS Appl. Mater. Interfaces* **12**, 21368–21385 (2020).

42. Deng, S. *et al.* Manipulation of an ionic and electronic conductive interface for highly-stable high-voltage cathodes. *Nano Energy* **65**, 103988 (2019).

43. Shu, Y. *et al.* Synergistic effect of surface plane and particle sizes on the electrochemical performance of LiNi0.5Mn1.5O4 cathode material via a facile calcination process. *J. Power Sources* **433**, 226708 (2019).

44. Kiziltas-Yavuz, N. *et al.* Enhancement of electrochemical performance by simultaneous substitution of Ni and Mn with Fe in Ni-Mn spinel cathodes for Li-ion batteries. *J. Power Sources* **327**, 507–518 (2016).

45. Gruber, P. W. *et al.* Global lithium availability: A constraint for electric vehicles? *J. Ind. Ecol.* **15**, 760–775 (2011).

46. *Lithium statistics and information*. *U.S. Geological Survey,2020* vol. 53 (2020).

47. McKinsey&Company. Lithium and cobalt-a tale of two commodities. *McKinsey&Company Met. Min.* (2018).

48. Grant, A., Whittaker, M. & Aaron, C. Inside Tesla ' s Lithium Clay Salt Extraction Process. *Jade Cove Partners* https://www.jadecove.com/research/teslasaltclay.

49. van den Brink, S., Kleijn, R., Sprecher, B. & Tukker, A. Identifying supply risks by mapping the cobalt supply chain. *Resour. Conserv. Recycl.* **156**, 104743 (2020).

50. Fu, X. *et al.* Perspectives on Cobalt Supply through 2030 in the Face of Changing Demand. *Environ. Sci. Technol.* **54**, 2985–2993 (2020).

51. Farjana, S. H., Huda, N., Mahmud, M. A. P. & Lang, C. A global life cycle assessment of manganese mining processes based on EcoInvent database. *Sci. Total Environ.* **688**, 1102–1111 (2019).

52. Jordans, F. Why automakers are joining the push against deep seabed mining. *CS Monitors* https://www.csmonitor.com/Environment/2021/0331/Why-automakers-are-joining-the-push-against-deep-seabed-mining (2021).

53. Carver, R. *et al.* A critical social perspective on deep sea mining: Lessons from the emergent industry in Japan. *Ocean Coast. Manag.* **193**, 105242 (2020).

54. U.S. Geological Survey. Commodity Statistics and Information.

https://www.usgs.gov/centers/nmic/commodity-statistics-and-information.

55. BGS. Uk and World Mineral datasets and statistics. https://www.bgs.ac.uk/datasets/uk-and-world-mineral-statistics-datasets/.

56. Speirs, J., Contestabile, M., Houari, Y. & Gross, R. The future of lithium availability for electric vehicle batteries. *Renew. Sustain. Energy Rev.* **35**, 183–193 (2014).

57. Narins, T. P. The battery business: Lithium availability and the growth of the global electric car industry. *Extr. Ind. Soc.* **4**, 321–328 (2017).

58. Vikström, H., Davidsson, S. & Höök, M. Lithium availability and future production outlooks. *Appl. Energy* **110**, 252–266 (2013).

59. Calisaya-Azpilcueta, D., Herrera-Leon, S., Lucay, F. A. & Cisternas, L. A. Assessment of the supply chain under uncertainty: The case of Lithium. *Minerals* **10**, 1–20 (2020).

60. Marcelo, A., Goffaux, N. & Hoffman, K. How clean can the nickel industry become ? *McKinsey insights* 1–6 (2020).

61. Habib, K., Hansdóttir, S. T. & Habib, H. Critical metals for electromobility: Global demand scenarios for passenger vehicles, 2015–2050. *Resour. Conserv. Recycl.* **154**, 104603 (2020).

62. *Climate Change 2014 Synthesis Report*. *IPCC* (2014).

63. Olivetti, E. A., Ceder, G., Gaustad, G. G. & Fu, X. Lithium-Ion Battery Supply Chain Considerations: Analysis of Potential Bottlenecks in Critical Metals. *Joule* **1**, 229–243 (2017).

64. Haynes, W. M. *CRC Handbook of Chemistry and Physics*. (CRC Press, 2016).

65. Perks, C. & Mudd, G. Titanium, zirconium resources and production: A state of the art literature review. *Ore Geol. Rev.* **107**, 629–646 (2019).

66. Song, Y., Wang, N. & Yu, A. Temporal and spatial evolution of global iron ore supply-demand and trade structure. *Resour. Policy* **64**, 101506 (2019).

67. Li, S. *et al.* Risk identification and evaluation of the long-term supply of manganese mines in China based on the VW-BGR method. *Sustain.* **11**, 1–23 (2019).

68. LaRocca, G. M. Working Paper ID-067 Global Value Chains : Lithium in Lithium-ion Batteries for Electric Vehicles U . S . international Trade Commission Global Value Chains : Vehicles. (2020).

69. Gulley, A. L., McCullough, E. A. & Shedd, K. B. China's domestic and foreign influence in the global cobalt supply chain. *Resour. Policy* **62**, 317–323 (2019).

70. McKinsey. Has South East Asia taken a firm grip on the nickel value chain ? 2019 (2021).

71. Tesla partners with nickel mine amid shortage fears. *BBC News* https://www.bbc.co.uk/news/business-56288781 (2021).

72. Helbig, C., Bradshaw, A. M., Wietschel, L., Thorenz, A. & Tuma, A. Supply risks associated with lithium-ion battery materials. *J. Clean. Prod.* **172**, 274–286 (2018).

73. Ciez, R. E. & Whitacre, J. F. Examining different recycling processes for lithium-ion batteries. *Nat. Sustain.* **2**, 148–156 (2019).

74. Roberto Sifon-arevalo. Why lithium has turned from gold to dust for investors. *S&P Global* https://www.spglobal.com/en/research-insights/articles/why-lithium-has-turned-from-gold-to-dust-for-investors (2019).

75. Babbitt, C. W. Sustainability perspectives on lithium-ion batteries. *Clean Technol. Environ. Policy* **22**, 1213–1214 (2020).

76. Cobalt. *Trading Economics* https://tradingeconomics.com/commodity/cobalt (2021).

77. Nickel. *Trading Economics* https://tradingeconomics.com/commodity/nickel (2021).

78. Lithium. *Trading Economics* https://tradingeconomics.com/commodity/lithium (2021).

79. Sovacool, B. K., Hook, A., Martiskainen, M., Brock, A. & Turnheim, B. The decarbonisation divide: Contextualizing landscapes of low-carbon exploitation and toxicity in Africa. *Glob. Environ. Chang.* **60**, 102028 (2020).

80. May, I. V., Kleyn, S. V. & Vekovshinina, S. A. Assessment of impact of accumulated environmental damage to the quality of soil, surface and groundwater, agricultural products resulted from the mining enterprise. *IOP Conf. Ser. Earth Environ. Sci.* **315**, (2019).

81. Thies, C., Kieckhäfer, K., Spengler, T. S. & Sodhi, M. S. Assessment of social sustainability hotspots in the supply chain of lithium-ion batteries. *Procedia CIRP* **80**, 292–297 (2019).

82. Gifford, S. Building a Responsible Cobalt Supply Chain. *Faraday Insights* (2019).

83. Sonter, L. J., Dade, M. C., Watson, J. E. M. & Valenta, R. K. Renewable energy production will exacerbate mining threats to biodiversity. *Nat. Commun.* **11**, 6–11

(2020).

84. Wang, S. Metal Mining for Lithium-ion Battery Production of Electric Vehicles and Its Impact on Global Land Use : A Review. 21–31 (2020).

85. Lèbre, É. *et al.* The social and environmental complexities of extracting energy transition metals. *Nat. Commun.* **11**, 1–8 (2020).

86. Farjana, S. H., Huda, N., Parvez Mahmud, M. A. & Saidur, R. A review on the impact of mining and mineral processing industries through life cycle assessment. *J. Clean. Prod.* **231**, 1200–1217 (2019).

87. Kelly, J. C., Dai, Q. & Wang, M. Globally regional life cycle analysis of automotive lithium-ion nickel manganese cobalt batteries. *Mitig. Adapt. Strateg. Glob. Chang.* 371–396 (2019) doi:10.1007/s11027-019-09869-2.

88. Kallitsis, E., Korre, A., Kelsall, G., Kupfersberger, M. & Nie, Z. Environmental life cycle assessment of the production in China of lithium-ion batteries with nickel-cobalt-manganese cathodes utilising novel electrode chemistries. *J. Clean. Prod.* **254**, 120067 (2020).

89. Titanium Dioxide (TiO2) Production and Manufacturing Process. *Independant Commodity Intellegence Services (ICIS)*.

90. Lee, R. & Gifford, S. The importance of coherent regulatory and policy strategies for the recycling of EV batteries. *Faraday Insights* 8 (2020).

91. Richa, K., Babbitt, C. W. & Gaustad, G. Eco-Efficiency Analysis of a Lithium-Ion Battery Waste Hierarchy Inspired by Circular Economy. *J. Ind. Ecol.* **21**, 715–730 (2017).

92. Chen, P. *et al.* Reviving Aged Lithium-Ion Batteries and Prolonging their Cycle Life by Sinusoidal Waveform Charging Strategy. *Batter. Supercaps* **2**, 673–677 (2019).

93. Harper, G. *et al.* Recycling lithium-ion batteries from electric vehicles. *Nature* **575**, 75–86 (2019).

94. Garole, D. J. *et al.* Recycle, Recover and Repurpose Strategy of Spent Li-ion Batteries and Catalysts: Current Status and Future Opportunities. *ChemSusChem* **13**, 3079–3100 (2020).

95. Nordelöf, A. *et al.* Methodological approaches to end-of-life modelling in life cycle assessments of lithium-ion batteries. *Batteries* **5**, (2019).

96. gov.uk. Waste batteries: producer responsibility. https://www.gov.uk/guidance/waste-batteries-producer-responsibility#:~:text=It's illegal to send waste,sale on the UK market. (2021).

97. Moore, D. Lithium-ion battery waste fires costing UK over 150m a year. *Circular* https://www.circularonline.co.uk/news/lithium-ion-battery-waste-fires-costing-uk-over-150m-a-year/ (2021).

98. Gaines, L., Richa, K. & Spangenberger, J. Key issues for Li-ion battery recycling. *MRS Energy Sustain.* **5**, 1–14 (2018).

99. Piątek, J. *et al.* Sustainable Li-Ion Batteries: Chemistry and Recycling. *Adv. Energy Mater.* **2003456**, (2020).

100. Zhang, X. *et al.* Toward sustainable and systematic recycling of spent rechargeable batteries. *Chem. Soc. Rev.* **47**, 7239–7302 (2018).

101. Gaines, L. The future of automotive lithium-ion battery recycling: Charting a sustainable course. *Sustain. Mater. Technol.* **1**, 2–7 (2014).

102. Mayyas, A., Steward, D. & Mann, M. The case for recycling: Overview and challenges in the material supply chain for automotive li-ion batteries. *Sustain. Mater. Technol.* **19**, e00087 (2019).

103. Steward, D., Mayyas, A. & Mann, M. Economics and challenges of Li-ion battery recycling from end-of-life vehicles. *Procedia Manuf.* **33**, 272–279 (2019).

104. Fujita, T. *et al.* Reduction, reuse and recycle of spent Li-ion batteries for automobiles: A review. *Int. J. Miner. Metall. Mater.* **28**, 179–192 (2021).

105. Winslow, K. M., Laux, S. J. & Townsend, T. G. A review on the growing concern and potential management strategies of waste lithium-ion batteries. *Resour. Conserv. Recycl.* **129**, 263–277 (2018).

106. Guo, X., Zhang, J. & Tian, Q. Modeling the potential impact of future lithium recycling on lithium demand in China: A dynamic SFA approach. *Renew. Sustain. Energy Rev.* 110461 (2020) doi:10.1016/j.rser.2020.110461.

107. Kader, Z. A., Marshall, A. & Kennedy, J. A review on sustainable recycling technologies for lithium-ion batteries. *Emergent Mater.* 725–735 (2021) doi:10.1007/s42247-021-00201-w.

108. Huang, B., Pan, Z., Su, X. & An, L. Recycling of lithium-ion batteries: Recent advances and perspectives. *J. Power Sources* **399**, 274–286 (2018).

109. Liu, C., Lin, J., Cao, H., Zhang, Y. & Sun, Z. Recycling of spent lithium-ion batteries in view of lithium recovery: A critical review. *J. Clean. Prod.* **228**, 801–813 (2019).

110. Yao, Y. *et al.* Hydrometallurgical Processes for Recycling Spent Lithium-Ion Batteries: A Critical Review. *ACS Sustain. Chem. Eng.* **6**, 13611–13627 (2018).

111. Gao, H. *et al.* Efficient Direct Recycling of Degraded LiMn2O4Cathodes by One-Step Hydrothermal Relithiation. *ACS Appl. Mater. Interfaces* **12**, 51546–51554 (2020).

112. Xu, P. *et al.* Efficient Direct Recycling of Lithium-Ion Battery Cathodes by Targeted Healing. *Joule* **4**, 2609–2626 (2020).

113. Lander, L. *et al.* Financial viability of electric vehicle lithium-ion battery recycling. *iScience* **24**, 102787 (2021).

114. Tan, D. H. S., Xu, P. & Chen, Z. Enabling sustainable critical materials for battery storage through efficient recycling and improved design: A perspective. *MRS Energy Sustain.* **7**, 1–13 (2020).