http://pubs.acs.org/journal/aelccp

# Insights into the Critical Materials Supply Chain of the Battery Market for Enhanced Energy Security

Marm Dixit, Brett Witherspoon, Nitin Muralidharan, Matthew M. Mench, Chol-Bum M. Kweon, Yang-Kook Sun, and Ilias Belharouak\*



ABSTRACT: This paper delves into the critical materials supply chain of the battery market with an emphasis on longterm energy security. The study recognizes electric vehicle battery packs as reservoirs of "locked reserves" for extended periods, typically 10 years or more. A comprehensive understanding of material flows and end-of-life battery management is essential to establish a sustainable, durable, and secure domestic supply chain for lithium-ion batteries. In addressing these concerns, the paper introduces a metric designed to assess the "per mile" consumption of critical reserves called "Materials Per Gallon-Electric (MPGe)". The study emphasizes the immediate need for critical materials to meet the accelerated demand for large-scale electric vehicle adoption in the short



term. Furthermore, the paper also emphasizes the urgent need to advance recycling technologies to recover the critical mineral reserves "locked" in end-of-life battery packs.

ransitioning to an electrified transportation sector necessitates the production of battery storage capacities ranging from 1,000 to 2,000 GWh.<sup>1-</sup> Understanding and estimating material requirements throughout the lifetime of a large-scale electrified transport fleet is crucial to assessing the stability of the material supply chain over extended durations.<sup>5–8</sup> To facilitate effective communication among researchers, industry professionals, and policymakers, it is crucial to establish standard metrics for material usage.<sup>9</sup> Internal combustion engines (ICEs) primarily consume gasoline, and the prevalent metric for ICE vehicles is miles per gallon (MPG).<sup>10<sup>1</sup></sup> Currently, the US average for light-duty vehicles is 24.9 miles per gallon, with each vehicle traveling 12,000 miles in a year on average.<sup>11-13</sup> When used together, the average distance traveled and the miles per gallon metric, along with the number of vehicles in the fleet, provide a reasonable estimation of the total gasoline consumption in a year as well as over the lifetime of the ICE vehicles. With the metrics used here, it is estimated that a fleet of 100,000 ICE vehicles with a lifetime of 10 years would consume roughly 48 million gallons of gasoline, which would require approximately 100 million gallons of crude oil.<sup>14,15</sup> Supply chain metrics for ICE vehicles are easily estimated due to the straightforward nature of the fuel source and refining process, and the lack of

recycling options.<sup>16</sup> However, electric vehicle (EV) battery packs pose challenges due to complex material requirements, refining, processing, and recycling.<sup>17,18</sup> Herein, we propose using *materials per gallon-electric* as a metric for the battery supply chain. This metric aims to quantify the material requirements of EVs relative to the fuel consumption of ICE vehicles, offering a clearer understanding of the environmental impact and reserve utilization associated with each mode of transportation. Materials per gallon-electric (MPGe) estimates

Materials per gallon-electric (MPGe) estimates the amount of material required in an EV to offset the environmental impact of each gallon of fuel consumed by an ICE vehicle.

 Received:
 May 11, 2024

 Revised:
 June 24, 2024

 Accepted:
 June 27, 2024

 Published:
 July 10, 2024







Figure 1. Materials per Gallon-Electric (MPGe) and the Battery Supply Chain. (a) A schematic diagram explaining the materials per gallonelectric metric is proposed to evaluate the material requirement during the battery lifetime. (b) Sample calculations for the material requirement for the duration of one EV pack life. Absolute and equivalent materials per gallon-electric metric for one 260 mile pack are visualized in terms of electrode materials, precursors, and unrefined ores needed. Note that the equivalent MPGe metric is scaled by  $10^3$  to enable visualization. (c) Supply chain diagrams for NMC622 and LFP materials. The sankey diagram illustrates the flow of minerals from raw material extraction through ore processing, battery material processing, and finally into the production of EV battery packs. The diagram highlights the pathways and quantities of key minerals used in different stages of the supply chain. Raw material and ore required are estimated from processing and refining efficiencies reported in literature.<sup>19–24</sup> The reported metrics are in metric tons and normalized as materials per gallon-electric. The calculations assume 100,000 EVs, with a 260 mile battery pack, with a 10 year life averaging 12,000 miles/ year. See also the Python code in the Supporting Information for more plotting options.

the amount of material required in an EV to offset the environmental impact of each gallon of fuel consumed by an ICE vehicle. It considers several crucial factors, including the lifespan of the EV, the anticipated distance traveled over its lifetime, and the fuel efficiency of traditional ICE vehicles. The equation for evaluating the MPGe value is detailed in eq 1 and Figure 1a. The MPGe value identifies material reserves required to offset every gallon of fuel over a typical operational lifetime of the vehicle. A lower material per gallon value indicates a more reserve-efficient and environmentally friendly option. EVs with lower MPGe values require fewer materials to manufacture and operate over their lifespan. The materials per gallon-electric metric offers policy makers, consumers, and industry stakeholders a standardized tool for assessing the sustainability of EVs in comparison to traditional ICE vehicles. It facilitates informed decision-making by providing a holistic perspective on the environmental implications of transportation choices.

 $MPGe = \frac{Absolute Material Required}{Normalization Factor}$   $Normalization Factor = \frac{12000[miles/year] \times 10[year]}{24.9[miles/gallon]}$ (1)

We believe that this metric will help visualize material usage over the lifetime of electric vehicles and enable comparison with internal combustion engines (Figure 1a). Overall, MPGe forms a crucial comparison when viewed in the context of reserve availability, economics as well as environmental impact. One pertinent aspect that MPGe metric does not consider is the comprehensive assessment of energy requirements throughout the lifecycle of electric vehicle (EV) batteries, encompassing both manufacturing and material extraction processes as well as charging over a lifetime. Indeed, the energy expenditure associated with battery production and raw material extraction is a crucial factor in determining the overall environmental impact and reserve efficiency of EVs. We acknowledge the necessity of incorporating these energy costs into our analysis to provide a more holistic evaluation of EV sustainability. Additionally, the variability in the sources of energy used to charge EV batteries, highlights the importance of considering the carbon intensity of the electricity grid. While our study focuses on the material consumption aspect through the introduction of the MPGe metric, future iterations of our research could explore integrating energy consumption and carbon emissions data into the assessment framework to offer a more comprehensive understanding of the environmental implications of EV adoption.

An EV battery pack is typically specified in terms of range, power, and capacity, as well as a specific battery chemistry. This information allows us to estimate the required materials for a single battery pack, including cathode, anode, separator, electrolyte, and current collector, based on certain assumptions about the battery pack's architecture. Further, the raw materials needed for processing the required battery materials are backcalculated using proper process efficiencies.<sup>19</sup> Subsequently, the ore and refining requirements are estimated from the raw material needs based on mining and refining efficiencies for individual minerals. $^{20-24}$  The details of efficiencies used for each step are provided in the Supporting Information. Sample calculations for a standard NMC (LiNi<sub>x</sub>Mn<sub>y</sub>Co<sub>z</sub>O<sub>2</sub>)/graphite battery pack rated at 260 miles are shown in Figure 1a. The quantity of cathode and anode materials needed for 1 EV battery pack are approximately 130 and 84 kg, respectively. Similarly, the raw materials for Li, Ni, Co, and Mn are approximately 11, 48, 16, and 15 kg, corresponding to 15, 56, 21, and 20 kg of unrefined ores, respectively.<sup>20,22-27</sup> This absolute material requirement is normalized to the lifespan, expected distance traveled, and the fuel efficiency of ICE vehicles as described in Eqn 1 to provide the materials per gallon-electric metric. For one EV battery pack based on

For one EV battery pack based on NMC622/graphite chemistry, this corresponds to approximately 26.9 and 17.37 g of cathode and anode material per gallon over the lifetime of the EV, respectively.

NMC622/graphite chemistry, this corresponds to approximately 26.9 and 17.37 g of cathode and anode material per gallon over the lifetime of the EV, respectively. Similarly, the raw and unrefined ore can be normalized to obtain the materials per gallon-electric metric. This metric represents the amount of material consumed over a 10 year period in a single EV battery that travels 12,000 miles per year. The flow of materials for a fleet of 100,000 EVs powered by NMC and LFP cathode chemistries is shown via Sankey diagrams (Figure 1b,c). The Sankey plots are generated through an interactive Python code that is included in the Supporting Information, which can be used to create additional visualizations. It can be seen that 2.7 t of NMC cathode and 3.72 t of LFP cathode are required to offset every gallon of fuel that would be consumed for 100,000 ICE engines over their lifetime. The corresponding requirements for the raw material and unrefined ore are described in the Sankey diagram. Assuming current material costs associated with the precursors, the cost of raw materials for a single EV to offset a gallon of gasoline is around \$0.54/ MPGe for the NMC622 cathode. A spreadsheet that allows the calculation of cost breakdown for cathode per MPGe metric is

provided in the Supporting Information. To determine the amount of oil required to produce an equivalent gallon of gasoline, it is essential to consider the refining process. It was found that for every gallon of crude oil refined, approximately 0.466 gallons are obtained as gasoline, with the rest being other byproducts like DFO, paraffins, waxes, and asphalt.<sup>14</sup> Considering this, we estimated that roughly 2 gallons of crude oil is needed to produce a gallon of gasoline-including just the raw materials. Thus, the crude oil needed to get a gallon of gasoline is around \$4, based on current price trends. This cost analysis does not include processing or refining costs, but still shows that EVs are more cost-effective in terms of material utilization. High Coulombic efficiencies of batteries can reduce the overall material usage and corresponding costs, environmental impact, and reserve scarcity. Similarly, the usage of other reserves like Cu, Al, and carbon is tracked over the battery production cycle from unrefined ores to the finished battery pack (Figure 1b,c). We note here that Fe was not integrated into this analysis for LFP due to the general abundance of iron. However, the supplied visualization code can be leveraged to enable visualization of the Fe supply chain as needed. Visualization of the material flow pathways like the one described here is vital for assessing the long-term impact of material choice. We further discuss the need for design software for batteries and associated challenges and prospects in the Supporting Information.

Conventional Li-ion batteries use carbon-based materials as anode (graphite), polyolefin separators, organic solvent-based electrolyte, and oxide-type cathode materials. Of these, the anode, separator, and electrolyte do not pose an immediate threat in terms of raw material scarcity.<sup>29,30</sup> Cathode materials, which are typically Li-transition metal oxides, pose a crucial threat to energy security due to the coupled supply chain of raw minerals and the reserve scarcity thereof.<sup>31,32</sup> The total world reserves (Figure 2a) identified by the United States Geological Survey (USGS) show that Co has the lowest reserve of about 10 million metric tons, while Fe is the most abundant, with global reserves of about 10<sup>6</sup> million metric tons among the essential minerals of importance for batteries (Li, Ni, Co, Mn, Fe, Cu, and Al).<sup>26</sup> It is vital to track the expected growth in material requirement and the decrease in global reserves to identify potential scarcity in the coming years. Using predictions from NREL about the number of EVs that will be deployed by 2050, material usage predictions are performed based on the metrics identified previously for a single EV.<sup>33</sup> EV growth is expected to be accelerated in the near term due to policy decisions and the transition to the electrified fleet (until  $\sim 2030$ ), followed by a controlled growth period. This is clearly reflected in the projected material requirements (Figure 2b) for the critical battery components. Cathode materials pose a more crucial threat compared to other battery components in terms of supply chain resilience due to their reliance on critical materials. The increased demand for battery packs will manifest in the development of battery production facilities with TWh capacities.<sup>34</sup> Material requirements for Li, Ni, Co, and Mn, which are the primary raw minerals required for battery cathodes, are projected for gigafactories in Figure 2c, based on an expected growth in gigafactories deployed as shown in Figure 2d. Here, individual gigafactories are assumed to have a production capacity of 15 GWh, producing battery packs rated at 260 miles with NMC622-graphite chemistry. Currently, the USA has 5-10 gigafactories in the planning/execution stages, but a much



Figure 2. Reserve Availability and Predicted Consumptions. (a) Total mineral reserves for key battery components reported by USGS in 2024.<sup>27</sup> These metrics provide the economically viable, identified reserves as of 2024. These numbers may deviate based on usage and/or identification of new reserves, among other factors. (b) Predicted consumption of key battery materials for the next 50 years. The predictions are based on the numbers of EVs expected on the road in the next 25 years based on an earlier report.<sup>28</sup> The EVs are assumed to have a 260 mile pack with an NMC622-graphite chemistry. (c) Required mineral quantities as a function of established gigafactories. Each gigafactory is assumed to have an annual production of 15 GWh battery capacity. The batteries are assumed to be NMC622-graphite chemistry and (d) Typical battery capacities estimated to be deployed as a function of established gigafactories.

higher number of battery production facilities are required to ensure that EV demands are met to ensure domestic energy security. Material requirements are expected to grow a couple of orders of magnitude for moving from 15 GWh to >2 TWh. This entails the development and stabilization of a sustainable, domestic supply chain that can deliver the required raw materials to these production facilities within the coming decade.

We also carried out an analysis of critical material demand and reserve depletion as a function of EV growth (Figure 3) for two major cathode chemistries: NMC and NCA (Li- $Ni_rCo_vO_2$ ). The quantity of unrefined raw Li, Ni, Co, and Mn for the projected number of EVs was estimated as previously described. The impact of this consumption on the global mineral reserves was subsequently estimated. For this, a mining efficiency factor and market utilization factor were defined for each mineral based on available literature. The market utilization factor was defined as the fraction of the available reserves used in the battery industry. This enabled the translation of the total global reserves identified by USGS (Figure 2a) to a quantity that is anticipated to be available to the battery industry. Three different metrics of the mining efficiency and market penetration are assumed for each mineral, corresponding to the three curves for global reserves in Figure 3. The details regarding the metrics employed are available in the Supporting Information. Current state-of-theart metrics of mining efficiency are approximately 75-85%,

depending on the mineral, with market utilizations ranging from 15% for Mn to 50% for Ni. These figures reflect the present scenario, where the battery industry is one of many competing sectors for these minerals. However, it is important to recognize that the demand for batteries is projected to grow exponentially in the future, driven by the increasing adoption of electric vehicles and the expansion of renewable energy storage solutions. By 2050, we anticipate that the market utilization of critical minerals for batteries will be substantially higher than current levels. This shift is primarily due to the relatively slower growth rates of other sectors that use these minerals. As a result, the battery sector's share of total mineral consumption is expected to increase markedly. This adjustment underscores the critical role that the battery industry will play in the future supply chain of these essential minerals and highlights the importance of strategic planning and investment in mineral extraction and recycling technologies to meet the burgeoning demand. Li, Ni, and Mn do not show a palpable scarcity over the projected period under these assumptions. However, Co reserve drops to <0.1 million metric tons by 2040 if employing an NMC chemistry under these conditions, indicating a severe material scarcity (Figure 3a). NCA-type cathode material is projected to enable a longer use of the available Co- reserves (Figure 3b) owing to the lower Co stoichiometry in the cathode compositions. Improving the mining efficiencies and market utilization are clear avenues for improving the overall reserve utilization and depletion metrics



Figure 3. Reserve Availability and Predicted Consumptions for Cathode Components. Predicted consumption of key battery materials for the next 50 years. The predictions are based on the numbers of EVs expected on the road in the next 25 years based on an earlier report.<sup>28</sup> The EVs are assumed to have a 260 mile pack with (a) an NMC622-graphite chemistry and (b) an NCA-graphite chemistry. Three reserve depletion curves are based on different metrics of ore refining yield, and the fraction of refined minerals used in the battery market. The details for the different scenarios are provided in the Supporting Information.



Figure 4. (a) Typical synthesis route employed for LiFePO<sub>4</sub> synthesis and the corresponding cost and mass distributions for the key raw materials. (b) Cost predictions for LFP raw materials under scenarios where phosphoric acid and lithium carbonate prices show  $\pm 100\%$  change. Five scenarios are picked from this contour plot and are identified as A-E. (c) Cost of LFP in \$/kg for the five scenarios picked from the contour plot. For actual cost (case I): 0.5 × Raw Material cost is assumed to be the processing cost for converting raw material to LFP cathode. For actual cost (case II): 0.25 × Raw Material cost is assumed to be the processing cost for converting raw material to LFP cathode.

over the coming decades. In addition, significant work needs to be carried out in the development of critical material-free alternatives that are practically deployable. Indeed, Co scarcity is widely known in the community, and significant work is carried out on the development of Co lean and Co-free cathode materials.<sup>35–38</sup> Translating this work from academic and national lab research into practical systems will require close association with industry partners.

The demand for cobalt-free cathode materials has revitalized interest in lithium iron phosphate (LFP) as a crucial battery chemistry. This shift is driven by the goal of reducing reliance on scarce and expensive materials, thus ensuring a sustainable and ethical supply chain.<sup>4,39–41</sup> LFP batteries are highly regarded for their low cost, exceptional safety, stable cycling performance, and good multiplier performance.<sup>42</sup> Consequently, LFP batteries have significantly increased their market share over the past decade, rising from 6% in 2020 to 34% in 2022, with an expected further increase to 39% by 2024. The global LFP battery market was valued at \$17.54 billion in 2023 and is projected to reach \$48.95 billion by 2031. As a result, the LFP battery supply chain plays a pivotal role in the transition to more sustainable and ethical battery technologies, particularly in the automotive sector.

Figure 4 illustrates a typical synthesis route for lithium iron phosphate (LiFePO<sub>4</sub>) and the associated cost and mass distributions for key raw materials, as well as future costing scenarios. Industrial synthesis of LiFePO<sub>4</sub> often involves solidstate synthesis, recognized as a cost-effective and energyefficient method. This process typically uses small molecule organic acids, like citric acid, as a carbon precursor alongside standard lithium, iron, and phosphorus sources. Phosphoric acid represents over 50% of the raw material by mass, followed by approximately 30% for the iron source and 20% for the lithium source. However, the cost distribution is skewed, with the iron source accounting for less than 1% of the total raw material cost, phosphoric acid less than 10%, and most of the cost is attributed to the lithium source. To assess future cost scenarios, we evaluated the relative change in raw material costs with fluctuations in lithium carbonate (Li<sub>2</sub>CO<sub>3</sub>) and phosphoric acid  $(H_3PO_4)$  costs. Large swings  $(\pm 100\%)$  in Li<sub>2</sub>CO<sub>3</sub> and H<sub>3</sub>PO<sub>4</sub> costs could lead to raw material costs being double the base cost at higher prices while dropping to onetenth of the current base prices at lower costs. The raw material costs under different scenarios could range from approximately \$4/kg to \$8/kg for current pricing and potentially as low as \$1/kg under certain conditions. Considering that raw materials account for approximately 50% of the total cathode cost, with the remaining 50% attributed to processing costs, the actual cost of LFP was evaluated in two cases. Case I assumed processing costs to be 0.5 time the raw material cost, resulting in LFP cost ranging from \$2/kg to \$11/kg, with a baseline price of approximately \$6/kg. In Case II, assuming processing costs are 0.25 times the raw material cost, the actual cost of LFP could range from \$1/ kg to \$9/kg, with a baseline price of approximately \$5/kg. This

This analysis underscores the importance of understanding and optimizing the LFP supply chain to ensure the continued affordability and sustainability of LFP batteries, particularly amidst fluctuating raw material costs.

analysis underscores the importance of understanding and optimizing the LFP supply chain to ensure the continued affordability and sustainability of LFP batteries, particularly amidst fluctuating raw material costs.

A concurrent issue with the rise in the adoption of electric vehicles and the generation of TWh-scale battery systems is the challenge of dealing with the batteries at their end-of-life.<sup>43–48</sup> The number of spent Li-ion batteries (LIBs) is expected to reach almost 800,000 units by 2025.49 End-of-life (EOL) battery packs from electric vehicles contain critical materials that must be captured to ensure economic, technological, and ecological viability. However, only a small percentage of EOL battery packs are currently being recycled (~10-15%), with most ending up in landfills.<sup>50</sup> The recycling infrastructure for LIBs is still immature and technologically nascent, and economy of scale does not exist, making them economically unfeasible. For EV batteries specifically, there are three main pathways for EOL management. These include remanufacturing into highly functioning EV batteries, remanufacturing into stationary storage batteries, and recycling for critical materials recovery. These pathways have varying values, with the highest

value option being the remanufacturing of batteries into highly functioning EV batteries, followed by the remanufacturing of batteries into stationary storage batteries and recycling critical materials recovery. However, as competition from battery manufacturers increases, critical material concentrations decrease, and environmental regulations become more stringent, the scrap battery values on a per kWh basis are expected to decline, driving the evolution of more innovative remanufacturing and recycling processes.

Currently, there are three primary modalities through which battery critical materials are accessed from the EoL battery packs (Figure 5a).<sup>51</sup> Hydrometallurgical recycling involves the use of liquid solutions, typically acids or bases, to extract valuable metals from spent Lithium-ion batteries. The process includes several steps: (a) Collection and Sorting: Batteries are collected and sorted based on their types and compositions; (b) Leaching: The sorted batteries undergo a leaching process where they are submerged in a liquid solution. This solution helps dissolve and extract metals from the battery components; (c) Solvent Extraction: After leaching, solvent extraction is employed to separate and concentrate specific metals such as lithium, cobalt, and nickel. This step helps in achieving high purity of the extracted metals; (d) Precipitation and Crystallization: The extracted metals are then precipitated or crystallized from the solution, resulting in metal-rich compounds; (e) Refining: The obtained compounds undergo further refining processes to produce pure metals suitable for reuse in new battery manufacturing. This method offers high metal recovery rates. However, it requires significant energy input and may generate large volumes of wastewater. Pyrometallurgical recycling, on the other hand, involves hightemperature processes to recover metals from the battery materials. The key steps for this processing method include: (a) Collection and Crushing: Batteries are collected and crushed to reduce them to a manageable size; (b) Smelting or Direct Melting: The crushed batteries are subjected to high temperatures through smelting or direct melting. This process results in the separation of metals from other battery components; (c) Separation and Refining: After melting, the molten material is separated into different layers based on the densities of the components. Metals are then further refined to achieve the desired purity; and (d) Casting: The refined metals are cast into ingots or other forms for reuse in manufacturing new batteries. This method is energy-intensive but can handle various types of battery chemistries. Both these technologies are commercially prevalent; however, they possess drawbacks, which include the need for shredding of the batteries, high energy consumption, and significant waste and greenhouse gas emissions. Further, the recovery efficiencies of these processes are low, in the range of 15-20%. In comparison, direct recycling focuses on reusing battery materials without extensive dismantling. This process involves: (a) Collection and Disassembly: Batteries are collected and disassembled into their individual components. This may include separating the cathode, anode, and electrolyte; (b) Sorting: The separated components are sorted based on their types and conditions; (c) Reassembly or Reuse: The sorted components are either directly reused in new battery manufacturing or reassembled into functional battery modules; and (d) Quality Checks: Before reuse, the components or assembled batteries undergo quality checks to ensure performance and safety standards are met. This approach mitigates the need for shredding of the batteries, as well as extensive chemical processing, leading to



Figure 5. Importance of Recycling to Sustain Battery Production. (a) Avenues for battery recycling and the resulting material from each process. (b) Materials per gallon-electric metrics for battery electrodes, precursors, and unrefined ores under four different recycling scenarios as described. The colors represent different recycling scenarios as described in the legend. The calculations assume 100,000 EVs, with a 260 mile battery pack, with a 10 year life averaging 12,000 miles/year. Projections for materials required for meeting EV demand assuming no current and high recycling efficiencies for (c) cathode, (d) cobalt mineral, and (e) nickel minerals. The predictions are based on the numbers of EVs expected on the road in the next 25 years based on an earlier report.<sup>28</sup> The EVs are assumed to have a 260 mile pack with an NMC622-graphite chemistry. Refer to Supporting Information for Recycling Numbers.

potential cost savings and reduced environmental impact. Further, the typical efficiencies for direct recycling methods are typically higher at  $\sim$ 30%, which is significantly improved from the hydro- and pyrometallurgical approaches.

The impact of recycling approaches and their associated efficiencies are evaluated in Figure 5b. Depending on the modality of recycling used, we obtain either (1) cathode material (direct recycling); (2) cathode precursors (hydrometallurgical recycling) or (3) unrefined ore (pyrometallurgical recycling). Correspondingly in the figure, the cathode section assumes a direct recycling approach with three different efficiencies; the battery precursor section assumes hydrometallurgical recycling with three different efficiencies, while the unrefined ore section considers a pyrometallurgical approach with three different efficiencies as outlined. Considering the case of 100,000 EVs, with a 260 mile battery pack and a 10 year life averaging 12000 miles/year, the amount of cathode required in the MPGe metric is ~2.7 MT. However, considering direct recycling at 80% efficiency, this requirement for virgin materials can be brought down to ~0.5 MT. Similar trends are seen overall for the precursor requirements as well as unrefined ores. Note that the recycling numbers assume that we have spent batteries corresponding to 100,000 EVs, all of which are undergoing recycling under a specific pathway. The scenario might be significantly more complex, involving multiple recycling streams and substantial variations in the end-of-life battery supply. Similarly, the projections over the years for cathode materials and Ni and Co reserves go significantly lower as the recycling efficiencies are improved while all the materials from EoL batteries are recycled (Figure 5C). Overall, these calculations underscore the paramount need for recycling technologies in ensuring a safe supply chain of lithium-ion battery technology. There is an urgent need to

There is an urgent need to increase the overall quantity of lithium-ion batteries being recycled while at the same time developing technologies that push the efficiency of recycling technologies to higher values.

increase the overall quantity of lithium-ion batteries being recycled while at the same time developing technologies that push the efficiency of recycling technologies to higher values.

In addition to its role in assessing material requirements and recycling potential, the MPGe metric offers several benefits that make it a valuable tool for evaluating the sustainability of different battery chemistries. One of the primary advantages of the MPGe metric is its ability to provide a standardized measure of material intensity relative to energy output, which facilitates direct comparisons across various battery technologies. This standardized approach allows researchers and industry stakeholders to easily identify which chemistries are more reserve-efficient and have lower environmental impacts, thereby informing decisions on material selection and battery design. The MPGe metric is particularly useful in scenarios where the trade-offs between material use and energy storage efficiency need to be clearly understood. For instance, in comparing high-nickel cathodes such as NCA and NMC622 with more abundant alternatives like LFP, the MPGe metric highlights the material efficiency of each option relative to the energy they deliver. This can guide investment in research and development toward chemistries that optimize both performance and sustainability. Moreover, the metric can be integrated into lifecycle assessments and techno-economic analyses to provide a comprehensive view of the environmental and economic impacts of battery production and recycling. Despite its advantages, the MPGe metric also has limitations that must be acknowledged. One significant limitation is that it focuses primarily on the material input side and does not account for the entire lifecycle impacts, such as energy consumption during manufacturing, operational efficiency, and end-of-life recycling processes. Additionally, while MPGe provides a useful measure of material intensity, it does not directly address other important factors like supply chain vulnerabilities, geopolitical risks associated with critical materials, or the social impacts of mining and material extraction. To address these limitations, it is essential to use the MPGe metric in conjunction with other evaluation tools and metrics that capture the full spectrum of sustainability concerns. By combining MPGe with comprehensive lifecycle assessments, supply chain risk analyses, and social impact evaluations, stakeholders can achieve a more holistic understanding of the trade-offs and benefits associated with different battery chemistries. This integrated approach ensures that decisions are informed by a balanced consideration of material efficiency, environmental impact, and broader sustainability goals. The focus of this manuscript is to examine the initial materials requirements for various battery chemistries. However, it is important to also take into account the total cost of ownership (TCO) over the lifespan of the battery cell/pack. This should include potential benefits from recycling at the end of life (EoL), which can significantly impact the overall economic feasibility of different battery chemistries. Recycling is particularly relevant for NMC and NCA chemistries, which are more frequently recycled compared to LFP. This variance in recycling potential can have an impact on the price balance between these chemistries. Although this manuscript does not go into a detailed technoeconomic analysis of recycling, preliminary assessments can be carried out using models like EverBatt, which are available in the existing literature.

A typical EV battery pack symbolizes "locked reserves" in the market over a duration of 10 years or more. Understanding the material flow, requirements, and management needed for EoL systems is crucial to achieving a sustainable, durable, and safe domestic supply chain for lithium-ion batteries. In this perspective, we highlighted a metric that provides a valuable gauge to assess the "per mile" consumption of the critical reserves that are used in a typical EV battery pack (viz. Co, Ni, Mn). We also supported this with open-source codes that enable battery design and evaluation of the associated supply chains corresponding to these systems. For large-scale adoption of EVs, we notice a strong need for critical materials in the short term to keep up with the accelerated demands. In addition to this, we highlight the imminent and urgent need for ramping up recycling technologies to recover these "locked" critical mineral reserves from within the EoL battery packs. Our analysis suggests a strong need for the development of new, salient technologies that push the efficiency limits of the current recycling process to achieve a sustainable battery supply chain.

# ASSOCIATED CONTENT

## **③** Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsenergylett.4c01300.

Discussion on open source battery design and supply chain toolkits (PDF) Cost Calculations (XLSX) Recycling Calculations (XLSX) Supply Chain Calculations (XLSX) Python resources to enable visualization (ZIP)

## AUTHOR INFORMATION

### **Corresponding Author**

Ilias Belharouak – Electrification and Energy Infrastructures Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830, United States; o orcid.org/0000-0002-3985-0278; Email: belharouaki@ornl.gov

#### Authors

- Marm Dixit Electrification and Energy Infrastructures Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830, United States; Orcid.org/0000-0002-9599-9288
- Brett Witherspoon Electrification and Energy Infrastructures Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830, United States
- Nitin Muralidharan Electrification and Energy Infrastructures Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830, United States; Department of Chemical Engineering, Indian Institute of Technology Madras, Chennai 600036, India
- Matthew M. Mench Department of Mechanical, Aerospace, and Biomedical Engineering, University of Tennessee, Knoxville, Tennessee 37996, United States
- **Chol-Bum M. Kweon** Computational and Information Sciences Directorate, Combat Capabilities Development Command Army Research Laboratory, Aberdeen Proving Ground, Maryland 21005, United States
- Yang-Kook Sun Department of Energy Engineering, Hanyang University, Seoul 04763, Republic of Korea; orcid.org/0000-0002-0117-0170

Complete contact information is available at: https://pubs.acs.org/10.1021/acsenergylett.4c01300

## **Author Contributions**

Marm Dixit: Conceptualization, Data curation, Formal Analysis, Investigation, Methodology, Writing-original draft,

## ACS Energy Letters

Writing-review and editing. Brett Witherspoon: Formal Analysis, Investigation, Methodology, Writing-original draft, Writing-review and editing. Nitin Muralidharan: Formal Analysis, Investigation, Methodology. Yang Kook Sun: Formal Analysis, Investigation, Methodology, Writing-original draft, Writing-review and editing. Ilias Belharouak: Conceptualization, Writing-review and editing, Funding acquisition, Resources, Supervision. Matthew Mench: review and editing, Funding acquisition, Chol-Bum M. Kweon: review and editing, Funding acquisition, Resources, Supervision.

#### Notes

The authors declare no competing financial interest.

#### Biographies

**Marm Dixit** is a R&D Staff Scientist and a previous Weinberg Distinguished Staff Fellow at the Oak Ridge National Laboratory. His research focus is solid-state and lithium metal batteries, lithium-ion batteries for eVTOL applications, advanced characterization as well as data science and analytics.

**Brett Witherspoon** is an Embedded Systems Hardware & Software Engineer in the Sensors and Electronics Group at ORNL. He specializes in electronics design and analog/digital signal processing for low-power embedded systems. Since joining ORNL in 2021 he has focused primarily on radiation monitoring systems and embedded neuromorphic systems.

Nitin Muralidharan is an assistant professor at Indian Institute of Technology Madras. He is a Material Scientist and Chemical Engineer with background in the area of next-generation energy storage technologies for Electric Vehicle (EV) and Electric Vertical Take-off and Lift (EVTOL) applications.

**Matthew M. Mench** is the Wayne T. Davis Dean's Chair, Chancellor's Professor, and Condra Chair at the Tickle College of Engineering at University of Tennessee Knoxville. His research interests span multiphase transport phenomena, advanced diagnostics, sensors, and modeling of power conversion and storage systems.

**Chol-Bum M. Kweon** is the Team Leader of Power Generation Research Team at the US Army Research Laboratory. He has published more than 60 technical and journal papers and 31 intellectual properties. He also serves as a Technical POC and Committee Member in multiple DoD organizations and technical communities.

Yang-Kook Sun is presently professor of Energy Engineering at the Hanyang University in Seoul. His research focuses on design, synthesis and structural analysis of advanced electrochemical energy storage and conversion materials. He is author of more than 510 publications in peer-reviewed scientific journals, and 340 registered and applied patents.

Ilias Belharouak is a Corporate Fellow and Head of the Electrification Section at the Oak Ridge National Laboratory and Professor at the Bredesen Center for Interdisciplinary Research and Graduate Education of the University of Tennessee Knoxville.

#### ACKNOWLEDGMENTS

This research at Oak Ridge National Laboratory, managed by UT Battelle, LLC, for the US Department of Energy under contract DE-AC05-00OR22725, was sponsored by the US Army DEVCOM Army Research Laboratory and was accomplished under Support Agreement 2371-Z469-22. The views and conclusions contained in this document are those of the authors and should not be interpreted as representing the official policies, either expressed or implied, of the DEVCOM Army Research Laboratory or the U.S. Government. This manuscript has been authored in part by UT-Battelle, LLC, under contract DE-AC05-00OR22725 with the US Department of Energy (DOE). The publisher, by accepting the article for publication, acknowledges that the US government retains a nonexclusive, paid-up, irrevocable, worldwide license to publish or reproduce the published form of this manuscript, or allow others to do so, for US government purposes. DOE will provide public access to these results of federally sponsored research in accordance with the DOE Public Access Plan (https://www.energy.gov/doe-public-access-plan).

## REFERENCES

(1) Liu, J.; Xiao, J.; Yang, J.; Wang, W.; Shao, Y.; Liu, P.; Whittingham, M. S. The TWh challenge: Next generation batteries for energy storage and electric vehicles. *Next Energy* **2023**, *1* (1), 100015.

(2) Kurtz, S. R.; Leilaeioun, A.; King, R. R.; Peters, I. M.; Heben, M. J.; Metzger, W. K.; Haegel, N. M. Revisiting the Terawatt Challenge. *MRS Bull.* **2020**, *45* (3), 159–164.

(3) Masias, A.; Marcicki, J.; Paxton, W. A. Opportunities and Challenges of Lithium Ion Batteries in Automotive Applications. *ACS Energy Letters* **2021**, *6* (2), 621–630.

(4) Sun, Y.-K. Direction for Development of Next-Generation Lithium-Ion Batteries. ACS Energy Letters **2017**, 2 (12), 2694–2695.

(5) Huang, Y.; Li, J. Key Challenges for Grid-Scale Lithium-Ion Battery Energy Storage. Adv. Energy Mater. 2022, 12 (48), 2202197.
(6) Vesborg, P. C. K.; Jaramillo, T. F. Addressing the terawatt challenge: scalability in the supply of chemical elements for renewable energy. RSC Adv. 2012, 2 (21), 7933-7947.

(7) Calderon, J. L.; Smith, N. M.; Bazilian, M. D.; Holley, E. Critical mineral demand estimates for low-carbon technologies: What do they tell us and how can they evolve? *Renewable and Sustainable Energy Reviews* **2024**, *189*, 113938.

(8) Watari, T.; Nansai, K.; Nakajima, K. Major metals demand, supply, and environmental impacts to 2100: A critical review. *Resources, Conservation and Recycling* **2021**, *164*, 105107.

(9) Larrick, R. P.; Soll, J. B.; Keeney, R. L. Designing better energy metrics for consumers. *Behavioral Science & Policy* **2015**, *1* (1), 63–75.

(10) Larrick, R. P.; Soll, J. B. The MPG Illusion. Science 2008, 320 (5883), 1593–1594.

(11) Sivak, M.; Tsimhoni, O. Fuel efficiency of vehicles on US roads: 1923–2006. *Energy Policy* **2009**, *37* (8), 3168–3170.

(12) 2023 EPA Automotive Trends Report; US Environmental Protection Agency, 2023.

(13) Hu, Z.; Mehrjardi, R. T.; Lai, L.; Ehsani, M. Optimal Hybridization of Conventional ICE Vehicles. *Eng.* **2021**, *2* (4), 592–607.

(14) King, C. W.; Webber, M. E. The Water Intensity of the Plugged-in Automotive Economy. *Environ. Sci. Technol.* 2008, 42 (12), 4305–4311.

(15) Bento, A. M.; Roth, K.; Zuo, Y. Vehicle Lifetime and Scrappage Behavior: Trends in the U.S. Used Car Market. *Energy Journal* **2018**, 39 (1), 159–184.

(16) Ghahremanlou, D.; Kubiak, W. Integrated bioethanol-gasoline supply chain evolved by changing US Government policies-model and algorithm. *International Journal of Operational Research* **2023**, *48* (2), 141–177.

(17) Cheng, A. L.; Fuchs, E. R.; Karplus, V. J.; Michalek, J. J. Electric vehicle battery chemistry affects supply chain disruption vulnerabilities. *Nat. Commun.* **2024**, *15* (1), 2143.

(18) Jagani, S.; Marsillac, E.; Hong, P. The Electric Vehicle Supply Chain Ecosystem: Changing Roles of Automotive Suppliers. *Sustainability* **2024**, *16* (4), 1570.

(19) Knehr, K.; Kubal, J.; Ahmed, S. Cost Analysis and Projections for US-Manufactured Automotive Lithium-ion Batteries; Argonne National Laboratory (ANL): Argonne, IL (United States), 2024.

(20) Locating and Estimating Air Emissions from Sources of Manganese; Environmental Protection Agency, 1985.

(21) Energy and Environmental Profile of the U.S. Mining Industry; US Department of Energy, 2002.

(22) Olafsdottir, A. H.; Sverdrup, H. U. Modelling Global Nickel Mining, Supply, Recycling, Stocks-in-Use and Price Under Different Resources and Demand Assumptions for 1850–2200. *Mining, Metallurgy & Exploration* **2021**, 38 (2), 819–840.

(23) Talens Peiró, L.; Villalba Méndez, G.; Ayres, R. U. Lithium: Sources, Production, Uses, and Recovery Outlook. *JOM* **2013**, *65* (8), 986–996.

(24) Das, J.; Kleiman, A.; Rehman, A. U.; Verma, R.; Young, M. H. The Cobalt Supply Chain and Environmental Life Cycle Impacts of Lithium-Ion Battery Energy Storage Systems. *Sustainability* **2024**, *16* (5), 1910.

(25) Davies, S. H.; Christensen, P.; Holberg, T.; Avelar, J.; Heidrich, O. Raw Materials and Recycling of Lithium-Ion Batteries. *Emerging Battery Technologies to Boost the Clean Energy Transition*; Springer: Cham, 2024; p 143.

(26) Schulz, K. J.; DeYoung, J. H., Jr.; Seal, R. R., II; Bradley, D. C. Critical mineral resources of the United States—An introduction; US Geological Survey, 2017.

(27) Minerals Yearbook, Vol. I, Metals and Minerals; USGS, 2024.

(28) *Global EV Outlook* 2024; International Energy Agency, Paris, 2024. (accessed 06/20/2024).https://www.iea.org/reports/global-ev-outlook-2024.

(29) Egbue, O.; Long, S.; Kim, S. D. Resource Availability and Implications for the Development of Plug-In Electric Vehicles. *Sustainability* **2022**, *14* (3), 1665.

(30) Olivetti, E. A.; Ceder, G.; Gaustad, G. G.; Fu, X. Lithium-ion battery supply chain considerations: analysis of potential bottlenecks in critical metals. *Joule* **2017**, *1* (2), 229–243.

(31) Murdock, B. E.; Toghill, K. E.; Tapia Ruiz, N. A perspective on the sustainability of cathode materials used in lithium ion batteries. *Adv. Energy Mater.* **2021**, *11* (39), 2102028.

(32) Yang, Z.; Huang, H.; Lin, F. Sustainable electric vehicle batteries for a sustainable world: perspectives on battery cathodes, environment, supply chain, manufacturing, life cycle, and policy. *Adv. Energy Mater.* **2022**, *12* (26), 2200383.

(33) Borlaug, B.; Ge, Y.; Lee, D.-Y.; Liu, Z.; Rose, E.; Spath Luhring, L.; St Louis-Sanchez, M.; Wood, E.; Yang, F. *Modeling Future Demand for EV Charging Infrastructure*; National Renewable Energy Laboratory (NREL): Golden, CO (United States), 2023.

(34) Moores, S. The global battery arms race: lithium-ion battery gigafactories and their supply chain. *Oxford Energy Forum*; Oxford Institute for Energy Studies, 2021; pp 26–30.

(35) Essehli, R.; Parejiya, A.; Muralidharan, N.; Jafta, C. J.; Amin, R.; Dixit, M. B.; Bai, Y.; Liu, J.; Belharouak, I. Hydrothermal synthesis of Co-free NMA cathodes for high performance Li-ion batteries. *J. Power Sources* **2022**, *545*, 231938.

(36) Li, W.; Lee, S.; Manthiram, A. High nickel NMA: a cobalt free alternative to NMC and NCA cathodes for lithium ion batteries. *Adv. Mater.* **2020**, 32 (33), 2002718.

(37) Muralidharan, N.; Essehli, R.; Hermann, R. P.; Amin, R.; Jafta, C.; Zhang, J.; Liu, J.; Du, Z.; Meyer, H. M., III; Self, E. Lithium Iron Aluminum Nickelate, LiNixFeyAlzO2—New Sustainable Cathodes for Next Generation Cobalt Free Li Ion Batteries. *Adv. Mater.* **2020**, 32 (34), 2002960.

(38) Muralidharan, N.; Self, E. C.; Dixit, M.; Du, Z.; Essehli, R.; Amin, R.; Nanda, J.; Belharouak, I. Next-Generation Cobalt-Free Cathodes - A Prospective Solution to the Battery Industry's Cobalt Problem. *Adv. Energy Mater.* **2022**, *12* (9), 2103050.

(39) Pozzato, G.; Li, X.; Lee, D.; Ko, J.; Onori, S. Accelerating the transition to cobalt-free batteries: a hybrid model for LiFePO4/ graphite chemistry. *npj Computational Materials* **2024**, *10* (1), 14.

(40) Sun, Y.-K. A Rising Tide of Co-Free Chemistries for Li-Ion Batteries. ACS Energy Letters 2022, 7 (5), 1774–1775.

(41) Voronina, N.; Sun, Y.-K.; Myung, S.-T. Co-Free Layered Cathode Materials for High Energy Density Lithium-Ion Batteries. *ACS Energy Letters* **2020**, *5* (6), 1814–1824.

(42) Yang, Y.; Liu, Z.; Zhang, J.; Chen, Y.; Wang, C. Economical and low-carbon regeneration of spent LiFePO4 materials by hydrothermal relithiation. *J. Alloys Compd.* **2023**, *947*, 169660.

(43) Ornes, S. How to recycle an EV battery. *Proc. Natl. Acad. Sci. U.* S. A. **2024**, *121* (5), e2400520121.

(44) Baum, Z. J.; Bird, R. E.; Yu, X.; Ma, J. Lithium-Ion Battery Recycling—Overview of Techniques and Trends. ACS Energy Letters 2022, 7 (2), 712–719.

(45) Bird, R.; Baum, Z. J.; Yu, X.; Ma, J. The Regulatory Environment for Lithium-Ion Battery Recycling. *ACS Energy Letters* **2022**, 7 (2), 736–740.

(46) Dunn, J.; Slattery, M.; Kendall, A.; Ambrose, H.; Shen, S. Circularity of Lithium-Ion Battery Materials in Electric Vehicles. *Environ. Sci. Technol.* **2021**, *55* (8), 5189–5198.

(47) Natarajan, S.; Aravindan, V. Recycling Strategies for Spent Li-Ion Battery Mixed Cathodes. *ACS Energy Letters* **2018**, 3 (9), 2101– 2103.

(48) Zhang, Q.; Gao, X.-W.; Liu, X.; Gu, Q.; Mu, J.; Luo, W.-B. Economical and Ecofriendly Lithium-Ion Battery Recycling: Material Flow and Energy Flow. ACS Sustainable Chem. Eng. 2024, 12 (7), 2511–2530.

(49) Shafique, M.; Rafiq, M.; Azam, A.; Luo, X. Material flow analysis for end-of-life lithium-ion batteries from battery electric vehicles in the USA and China. *Resources, Conservation and Recycling* **2022**, *178*, 106061.

(50) Velázquez-Martínez, O.; Valio, J.; Santasalo-Aarnio, A.; Reuter, M.; Serna-Guerrero, R. A critical review of lithium-ion battery recycling processes from a circular economy perspective. *Batteries* **2019**, 5 (4), 68.

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