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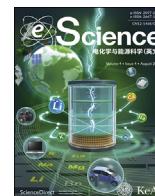
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Perspective

Opportunities and challenges in cathode development for non-lithium-ion batteries



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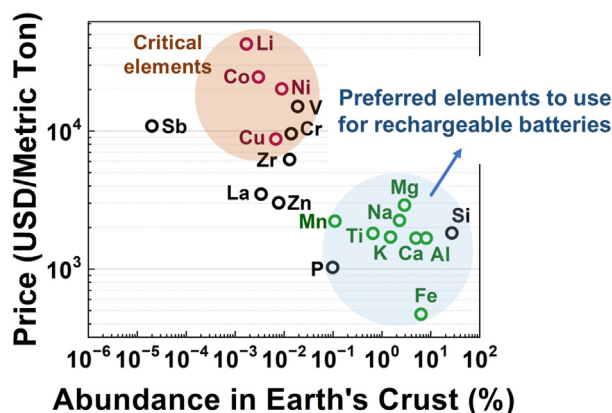
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HIGHLIGHTS

- This perspective discusses challenges in Li-ion batteries in the sustainability aspect.
- This perspective highlights the current status, important progress, and remaining challenges of the Li-alternatives.
- This perspective focuses on the selected non-lithium-ion, including Na-, K-, Mg-, and Ca-ion batteries.

GRAPHICAL ABSTRACT



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ABSTRACT

Lithium (Li)-ion batteries have stimulated the societal transformation to clean energy systems. This carry-on electricity is revolutionizing how society communicates, functions, and evolves efficiently by enabling mobile electronics, zero-emission electric vehicles, and stationary energy storage. In preparation for the sustainable energy future, however, there are growing concerns about depleting critical elements used in the Li technology (e.g., lithium, cobalt, and nickel), especially for large-scale applications that will accelerate the rate of elemental consumption. Various non-Li-based rechargeable batteries composed of earth-abundant elements, such as sodium, potassium, magnesium, and calcium, have been proposed and explored as alternative systems to promote sustainable development of energy storage. In this perspective, we discuss challenges in Li-ion batteries in the sustainability aspect and provide our opinions on the potential applications of non-Li-based batteries. We also highlight the current status, important progress, and remaining challenges of the Li-alternative technologies.

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1. Challenges of Li-ion batteries and opportunities of non-Li ion batteries

Energy storage technology catalyzes sustainability. Beyond portable electronics, lithium (Li)-ion batteries with improving energy storage capacity per weight and per volume (*i.e.*, specific energy and energy density, respectively) and reducing unit cost have been accelerating vehicle and grid electrification. As this societal transformation continues, the need for Li-ion batteries is increasing and will increase dramatically more in the near future.

Commodity prediction shown in Fig. 1a suggests that Li supplies may be sufficient for planned battery projects between now and 2025 [1]. However, rapidly growing demand for lithium carbonate equivalent (for instance, 500 thousand tons in 2022 to 2 million tons in 2030 [2,3], as shown in the inset) will outweigh its supply, assuming that the total Li reserve remains unchanged. As Li resources are highly localized in a few nations, the geographical Li production and utilization mismatch (Fig. 1b) will lead to geopolitical Li supply risks, and thereby price volatility.

Besides, modern Li-ion cathodes (*e.g.*, $\text{LiNi}_x\text{Co}_y\text{Mn}_z\text{O}_2$ where $x + y + z = 1$) necessitate cobalt (Co) and nickel (Ni) for efficient energy storage,

both of which are critical elements experiencing a pressing issue of depletion. The shortage of Co is already well documented in the literature [4]. With the increasing sales volume of electric vehicles (EVs), the worldwide Ni demand will also increase continuously, as shown in Fig. 1c [5]. While the largest portion of the current Ni production goes to the stainless steel industry, we expect that the Li-ion battery market will be soon the major contributor for Ni consumption. More critically, battery-grade Ni (> 99.8% purity) is difficult to produce, putting additional pressure on securing the resources for battery makers [6,7]. The statistical survey for the battery market forecasts that the known Ni reserve may run out as early as 2040 [6]. Not to mention, the price of critically essential elements for Li-ion batteries, including Li, Co, Ni, and Cu, has skyrocketed (Fig. 1d). Under these circumstances, the EV industry would consume almost all the currently produced Li-ion batteries preemptively, leaving very little room for new energy storage applications in the sustainability sector due to the lack of supply. Therefore, finding alternative technologies that promote broader dissemination of energy storage devices is urgently needed for a sustainable energy future.

In this respect, non-lithium-based rechargeable batteries made of abundant and low-cost elements shown in Fig. 1d (shaded in blue) have been extensively studied as alternative energy storage systems for various

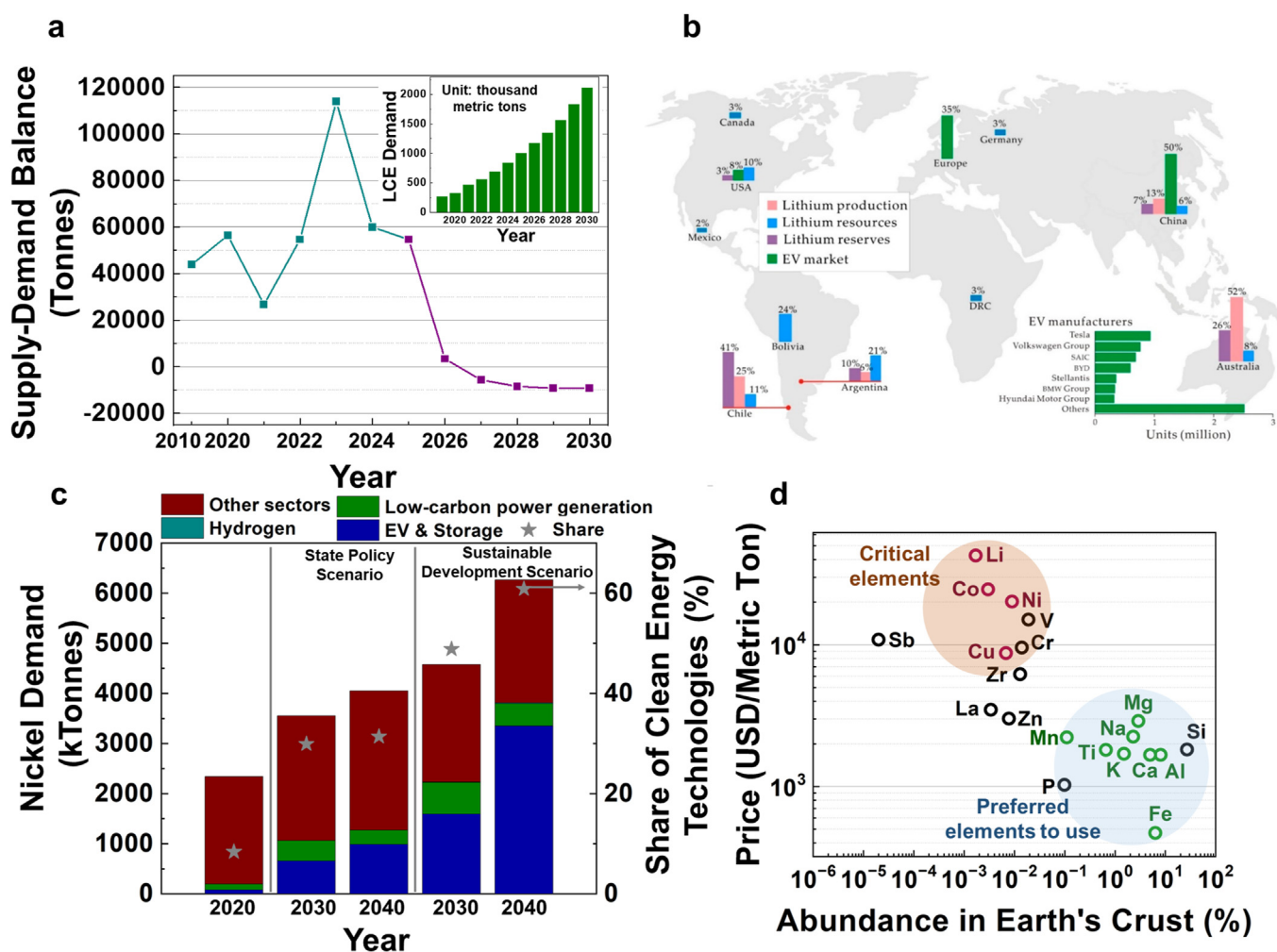


Fig. 1. (a) Supply-demand balance of the lithium carbonate equivalent (LCE) projected. (Inset: LCE demand projected). The data were obtained from online resources: [fastmarkets.com](https://www.fastmarkets.com) and [statista.com](https://www.statista.com). (Accessed September 2023) [1,2] (b) The availability of lithium and electric vehicle sales by countries in 2021. This figure presents round numbers of countries greater than 2% excluding the United States production. Adapted with permission under a Creative Commons Attribution (CC BY) license from Ref. [13] Copyright 2022 MDPI. (c) Projected nickel demands in different industry sectors. The data were obtained from online resource: [iea.org](https://www.iea.org) (Accessed September 2023) [5] (d) Abundance vs. Price of metal resources. The data were obtained from online resources: [metal.com](https://www.metal.com), [USGS.gov](https://www.usgs.gov), and <https://periodictable.com/Properties/A/CrustAbundance.an.html> (Accessed July 2023).

applications. These include sodium (Na), potassium (K), magnesium (Mg), and calcium (Ca)-ion batteries that have little, if not none, dependence on the critical elements [8–12]. It should be pointed out that we do not anticipate that one battery system will and can monopolize every energy storage market. Instead, different battery systems likely have different application domains given their intrinsically distinct electrochemical properties and technical maturity. For example, we believe Li-ion batteries will continue to dominate the long-range, high-performance EV market because of their high energy density. In contrast, the non-lithium technologies may penetrate other types of energy storage applications, including personal mobility, home backup power, and grid storage, all of which energy storage cost tends to matter more than energy density.

2. Alkali metal-ion systems: Na- and K-ion batteries

Energy storage that uses Na as a charge carrier, *i.e.*, Na-ion batteries, has a long and rich history in materials research. As alkali metals have common chemical characteristics, simple layered oxides have been considered a promising class of materials for Li- and Na-ion batteries [14]. In the early 1980s, almost right after layered LiCoO_2 came out for Li-ion cathodes, reversible Na intercalation into the layered oxide system was also demonstrated [15]. While the research momentum to advance Na storage materials had redirected to Li chemistry due to the rapid success of Li-ion batteries in the 1980s–2000s, the number of technical publications about Na-ion batteries started to regrow in the early 2000s because the need for low-cost batteries made of earth-abundant elements emerged [16].

Thermodynamically, Na-intercalating layered oxides undergo prominent, multiple phase transformations resulting from oxygen stacking change and Na/vacancy ordering upon Na extraction and reinsertion [17]. The complex structural evolutions result in multiple voltage steps and plateaus as a function of Na content. For most Na layered oxide cathodes, the charge process is limited at 4 V to avoid irreversible phase transformations that lead to abrupt capacity decay [16,17]. This voltage constraint often makes practical capacity achievable at ~60% (~140 mAh/g) of their theoretical values. Recent studies have demonstrated that judicious selection of the transition metal compositions can accommodate alternating multiple oxygen stackings and suppress the irreversible phase transformations, as a result of complex physicochemical interplays between Na, transition metals, and oxygen, stabilizing desodiated layered structures at high voltage [18–20].

Another interesting alkali metal element for intercalation electrochemistry is potassium (K). Materials research for K-ion batteries has been expanding since 2012 in search of low-cost Li alternatives [21]. However, practical cathodes to store K are yet to be developed due to materials design challenges. K ion is considerably heavier and larger than Li ion, and even than Na ion. Layered oxides that have been developed for Li and Na ions are unlikely suitable for reversibly hosting K ions due to

their compact unit cell volume. K occupancy in octahedral or prismatic sites in the layered structures will lead to substantial expansion of K slab spacing along the *c*-axis [21]. This makes neighboring K ions interact electrostatically with each other (*i.e.*, Coulombic repulsion) to a large extent as the in-plane $\text{K}^+ - \text{K}^+$ charge screening effect from surrounding oxygen becomes weak. Consequently, full K occupancy is unachievable in layered oxides that have no means to separate K ions physically and electrostatically [22,23]. In addition, the strong in-plane $\text{K}^+ - \text{K}^+$ Coulombic interaction makes significantly sloped voltage curves that limit the achievable capacity (< 140 mAh/g) and lower the average discharge voltage (< 3.0 V vs. K/K^+) [8].

Polyanionic materials that provide distinctive separation between alkali ions with excellent chemical stability may offer new design insights for K-ion batteries, as well as for Na-ion batteries. Fig. 2 compares molecular weights and theoretical specific capacities of layered ($\text{Co}^{3+/4+}$) and polyanionic ($\text{V}^{3+/4+}$) materials to store Li, Na, and K ions. The one-electron theoretical capacities of LiCoO_2 , NaCoO_2 , and KCoO_2 decrease as the molecular weight increases. With reference to LiCoO_2 , the capacity decreases by 14% for NaCoO_2 and 25% for KCoO_2 . If the polyanion cathode is considered, this difference reduces as the molecular weight of the cathodes originates mostly from the polyanionic group. With reference to LiVPO_4F , the theoretical capacities of NaVPO_4F and KVPO_4F decrease by 9% and 16%, respectively. This indicates that the weight disadvantage of K substitution for Li in polyanionic cathodes is not as significant as that in layered oxides. Moreover, many polyanionic K-ion cathodes and some polyanionic Na-ion cathodes can operate by nearly full transition metal redox, practically outperforming their layered counterparts that have limited redox activities, especially at high rates [22]. Fig. 3 shows the specific energy and energy density domains of a

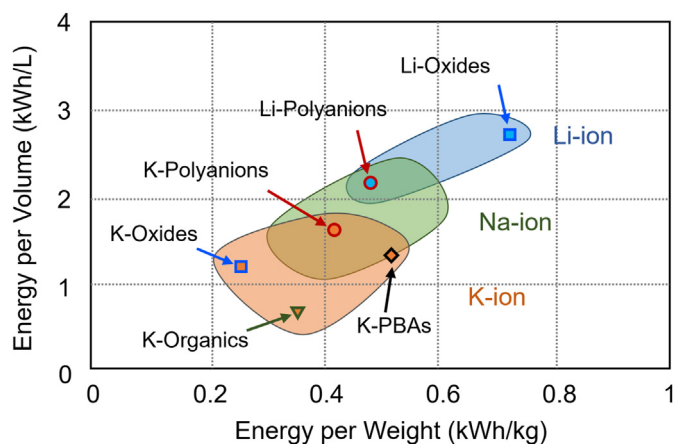


Fig. 3. Specific energy-energy density domains for different cathode chemistries.

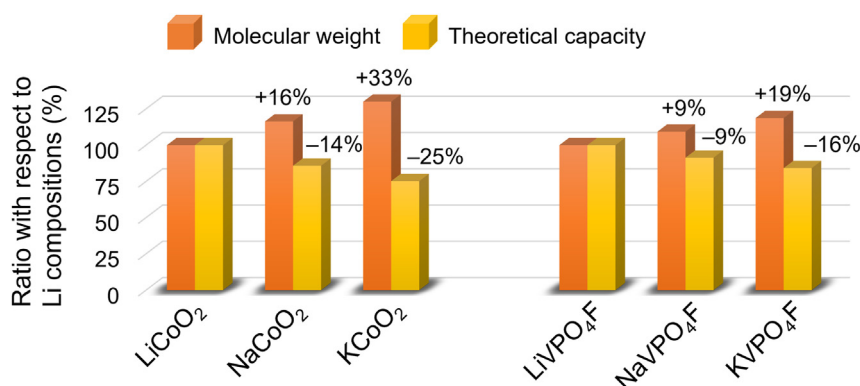


Fig. 2. Comparison of the relative molecular weights and specific capacities for Li, Na, and K compounds hosted by oxide and polyanionic frameworks.

few known Li, Na, and K-ion cathodes, as reported in the literature [21]. Indeed, polyanions can intercalate more K ions than layered oxides in practice, showing substantially higher specific energy and energy density.

Na-ion batteries are considered ready for commercialization. They have been expected to play a key role in stationary applications such as backup power and grid-level storage, in which mid-to-long-term energy storage cost (per cycle) is more important than the face value of energy density [24]. In this context, a number of Na-ion battery products have arrived and will arrive at the market for home energy solutions to store harvested solar power and/or replace diesel generators [25]. In addition to those applications, Na-based technologies nowadays see a new industrial horizon. As discussed earlier, the energy storage sector has been experiencing a pressing issue in allocating and distributing limited Li resources, yielding a new EV domain suitable for Na-ion batteries. The US Department of Energy suggests that an \$80/kWh battery pack, as manufactured, can make EVs cost-competitive [24,26]. Na-ion systems can meet this target if used for mid-range EVs [27]. Indeed, multiple automakers announced their business plans and roadmaps to manufacture the first Na-ion battery-powered EVs as early as 2023 [28,29]. Despite the sudden, significant expansion of the Na-ion battery market, we believe that collective effort to fundamentally tailor materials properties for Na-ion batteries still needs to continue in order to improve the energy density and cycling stability, strengthening their technological competitiveness.

It should be stressed that K-ion batteries have the unique potential to employ the graphite anode in combination with cathode materials made of earth-abundant elements. K storage materials that have open frameworks, such as Prussian blue analogs and organic compounds, can have reasonably high specific energy up to 500 Wh/kg but low energy density due to their large unit cells, as shown in Fig. 3. Their expected applications could be outdoor energy storage that is unrestricted by the unit volume. With balanced specific energy and energy density and chemical stability, polyanionic compounds, especially if manganese (Mn) or iron (Fe) redox can be utilized, may have a chance to develop practical K-ion batteries for indoor backup power. However, the K-based technology at present is technologically immature, and its estimated time of arrival in the energy storage market is unpredictable. In addition to materials design challenges, K chemistry at a cell level needs to address issues in electrode–electrolyte interfaces and interphases to enable reversible energy storage [30]. No clear understanding about developing electrochemically stable electrolytes exists, making the K-ion cell operate by poor coulombic efficiency and poor cyclability. Tailored approaches to optimize electrolytes that can promote facile and reversible K transport will greatly enhance the chance of K-ion batteries to disrupt the energy storage market. Efforts to stabilize the interfaces in K-ion batteries are highlighted elsewhere [31–33].

3. Multivalent ion systems: Mg- and Ca-ion batteries

Mg-ion batteries have several advantages over the Li-ion technology. Not to mention the abundance of Mg, critical transition metal elements such as Co and Ni are generally unnecessary in designing cathodes. More importantly, the Mg metal anode exhibits much higher volumetric capacity (3833 mAh/cm³) than Li metal (2061 mAh/cm³). Mg metal also exhibits high tolerance against detrimental dendrite growth, unlike Li metal. Because of these inherent advantages, Mg-ion batteries have been extensively studied since the first cathode composed of a chevrel phase (Mo₃S₄) was discovered [34]. Significant progress in understanding Mg intercalation reactions then led to the discovery of MgV₂O₄ and Mg(Mg_{0.5}V_{1.5})O₄ with exceptionally high discharge capacity of ~200 mAh/g [35–37].

However, most of the inorganic Mg-ion cathodes operate by relatively expensive vanadium (V) redox. To remain cost-effective per unit stored energy, it is required to expand the frontier of the chemical space of the

Mg storage materials to include Mn or Fe redox centers. Organic compounds have also been investigated as low-cost Mg-hosting cathode materials [38,39]. While implementing organic cathodes that exhibit high specific capacity may reduce the unit cost of Mg-ion batteries, their low material density tends to compromise energy density, one of the greatest strengths of Mg-ion batteries at a cell level.

A critical issue present in Mg-ion batteries is large cell polarization, *i.e.*, poor roundtrip efficiency, as commonly observed in emerging battery systems, in which cell-level understanding is lacking. Fig. 4 shows the range of voltage variation for a few known Mg-cathode materials. The top and bottom of the voltage variation extending from the average voltage value indicate the average charge and discharge voltage, respectively, required to obtain given capacity. For instance, MgCrMnO₄ has an average charge voltage at 2.75 V and an average discharge voltage of 1.8 V, making the average cell cycle voltage at 2.28 V [36]. If the cell redox reaction operates ideally, the average charge and discharge voltage should be identical. The 0.95 V difference, however, presents large voltage hysteresis, indicative of polarization. Thus, the length of the vertical voltage variation in Fig. 4 scales with the overall cell polarization. We found that most of the Mg-ion cathode materials have much larger polarization during charging and discharge than Li-ion cathode materials [35–38].

Although not as much investigated as Mg-ion batteries, Ca-ion batteries that leverage divalent Ca ions as a charge carrier are an interesting proposal for achieving high energy density. In this new system, the vanadium redox reactions to accommodate Ca extraction and reinsertion lead to large polarization, similar to that of the Mg system [40]. Diffusion-limited charge and discharge due to strong Coulombic interactions between the divalent cations and the host ionic crystals can be responsible for the large hysteresis in Mg- and Ca-ion batteries. However, the origin of polarization, whether it is intrinsic or can be reduced by engineering electrode configurations, is not yet fully deciphered and more in-depth studies to understand the energy storage mechanisms of Mg- and Ca-ion batteries are necessary.

The development of high-voltage electrolytes is also essential to the success of Mg- and Ca-ion batteries. Several nonaqueous electrolytes have demonstrated high voltage stability by cyclic and/or linear sweep voltammetry using a simple cell configuration of the current collector–electrolyte–metal anode [41–44]. However, the anodic stability of electrolytes (*i.e.*, electrochemical stability against oxidation at the cathode–electrolyte interface) also depends on the interphase formation with cathode composites. It should be also noted that Mg- and Ca-ion batteries often need elevated temperatures to deliver reasonable capacity due to kinetic limitation associated with the large voltage hysteresis. Therefore, rigorous electrochemical and thermal evaluations are required to better understand the compatibility of the electrolytes against highly oxidizing, charged cathode composites (including cathode active material, conductive carbon, and binder) for Mg- and Ca-ion batteries. Not to mention, the development of electrolytes that are compatible with the Mg and Ca metal anodes remains an important challenge.

Due to the technical immaturity of Mg- and Ca-ion batteries, predicting their future seems inappropriate. Yet, the multivalent system, as a whole, possesses clear advantages in achieving high energy density. If substantial development is made in materials design and cell engineering, we expect that they can have a potential market in personal short-range mobility applications, such as electric scooters and bicycles, and backup powers, in which unit cost and energy density are considered more important than specific energy due to affordability and space limitation.

4. Conclusion

This perspective article provides the past, present, and future of non-lithium-ion batteries, including monovalent Na and K and divalent Mg and Ca systems. Certainly, these are ongoing research topics, in which progress is being made in real time. Not to mention, other systems

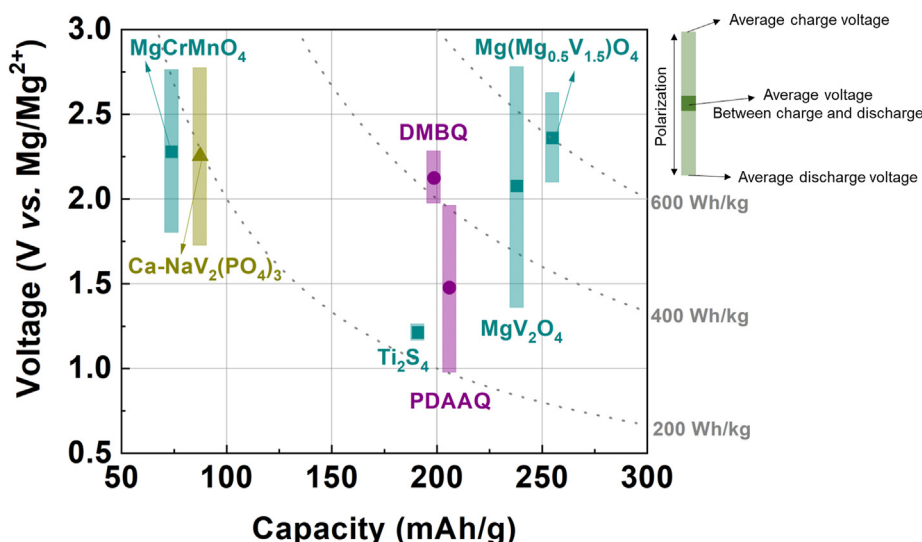


Fig. 4. Specific capacity vs. voltage plots of various Mg- and Ca-cathode materials. The bar represents the polarization between charging and discharging and darker marks show the average voltage. The data were obtained from literature [35–40,45].

leveraging Al and Zn ions as charge carriers can also be of interest to develop next-generation batteries beyond the Li-ion [46–48]. The Na-based system has shown the potential for commercialization and is close to market arrival, while the practical development of others faces challenges due to complex chemical and electrochemical problems. As each system has a unique advantage that can be distinguishable from each other, tailored research directions may lead to their success in market deployment. Na-ion batteries may be adapted for mid-range EVs and stationary storage systems, supplementing the Li technology. K-ion batteries would not compete with Li-ion directly but can have some market shares for small-scale, microgrid storage and home backup power if cyclability can be substantially improved. Mg and Ca can be used in applications where cost and energy density are the most critical requirements, but specific energy is not. Basically, each battery system can complement each other to offer green energy, shaping the sustainable energy future. Lastly, true technological innovation is often made by new materials development. In non-lithium-ion battery technologies, we emphasize that the development of high-performance electrode materials consisting of earth-abundant elements such as Fe and Mn will be an important pathway to enable greener and more sustainable battery systems.

Author contributions

Haeyeom Kim: Conceptualization, Funding acquisition, Visualization, Writing – original draft, Writing – review & editing. **Jae Chul Kim:** Conceptualization, Funding acquisition, Visualization, Writing – original draft, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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