

Commentary

Engineering current collectors for batteries with high specific energy

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INTRODUCTION

Batteries with high specific energy are attractive for a wide range of applications, such as the electrification of transport and portable electronics. Various approaches have been explored to increase the specific energy of batteries, such as developing new high-capacity electrode materials (e.g., high Ni-oxides, lithium metal, and sulfur) and increasing active material loading and density.^{1–4} Attaining key metrics for these components has led to higher specific energies, but the percentage of “dead weight” from metal current collectors (e.g., Cu for anodes and Al for cathodes) has increased as well. Therefore, making current collectors lighter becomes a promising approach to further increase battery specific energy, especially for lithium metal batteries with high specific energy. However, this strategy is often overlooked in literature. Here, we analyze the effect of current collector weight reduction on the specific energy of Li-(high Ni-oxide) and Li-S batteries, as well as other benefits and challenges. Our analysis focuses on pouch cells, given that it is a major form factor for vehicles and portable electronics. The analysis can be adapted to other form factors such as cylindrical cells, but different requirements between various types of cells should be taken into account, such as processing, mechanical strength, and tab configurations.

PAST EFFORTS IN CURRENT COLLECTOR WEIGHT REDUCTION

Current collector weight reduction has been explored since the early days of Li-ion battery development (Figure 1).

The limited literature shows that the typical thicknesses of Cu and Al current collectors were 20 and 18 μm , respectively in 1999, which accounted for ~19.3% and ~5.7% of the battery weight.⁵ The weight percentages reduced to 9.6% for Cu and 4.4% for Al in 2011,⁷ and in 2016, the thickness of Cu and Al were 10 and 15 μm , respectively.⁶ Currently, they are only 6 μm and 6.4% for Cu and 10 μm and 3% for Al in state-of-the-art.⁸ These values are based on cylindrical cells (1999 and 2018)^{5,8} and pouch cells (2011 and 2016), respectively.^{6,7} Thicker current collector foils were needed during the early years of battery production to compensate for fragility, defects, and non-uniform thickness. However, recent technological advancements have enabled thinner films with fewer defects and inhomogeneity and better mechanical properties, making thinner current collectors a possibility. For example, various additives can be used in electrodepositioning Cu to reduce defects inside. Moreover, better control in manufacturing processes, such as tension in slurry coating and winding, and compressive forces in calendaring, could help enable thinner current collectors in the future.

CURRENT COLLECTOR WEIGHT REDUCTION IN LITHIUM METAL BATTERY SYSTEMS

Although Cu and Al account for a lower portion of the battery weight nowadays, the fast development of high-capacity electrode materials might quickly reduce the weight of electrode materials and increase the contribution from the current collectors again. For example, a lithium anode with 6 mAh cm^{-2} capacity is only 1.5 mg cm^{-2} , only 50% of the weight of 6 μm Cu current collector (6/2 = 3 μm is considered as lithium is on both sides of Cu).

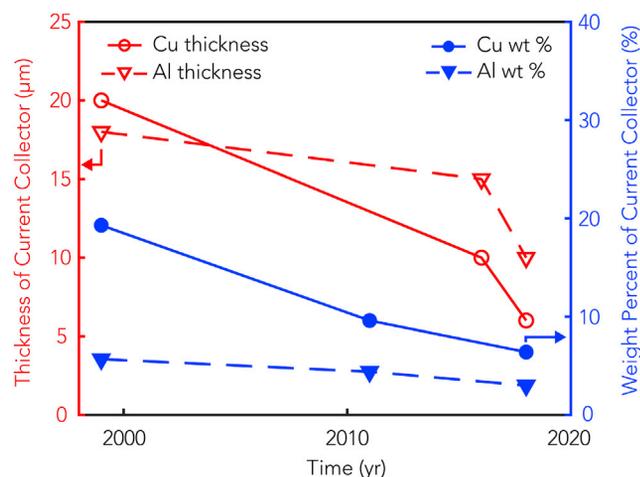


Figure 1. Past trends in battery current collectors

Thickness and weight percent of Cu and Al current collectors in conventional Li-ion batteries from 1999 to 2018. Data for 1999 and 2016 are based on the LiCoO₂-graphite chemistry,^{5,6} and data for 2011 and 2018 are based on the NCA-graphite chemistry.^{7,8}

Similarly, a sulfur cathode with 3 mAh cm⁻² capacity is only 3 mg cm⁻² based on 1,000 mAh g⁻¹ sulfur, merely double the weight of an Al current collector (10/2 = 5 μm considered). Therefore, weight reduction of current collectors enables an increase in the specific energy of these next-generation systems of 5%–20% (20–100 Wh kg⁻¹). Such benefits could release constraints on electrode materials and electrolyte to some extent (e.g., mass loading and electrolyte to electrode ratio). Therefore, understanding the effect of current collectors quantitatively is important for developing next-generation batteries. Two promising future technologies, Li/high Ni-oxide and Li/S systems, are used as representative systems in this commentary.

Figure 2A shows the dependence of specific energy on the thickness of Al and Cu current collectors in a Li/high Ni-oxide pouch cell based on a pouch cell model. The cathode loading is set as 3 mAh cm⁻² (15 mg cm⁻²), the N/P ratio (anode to cathode capacity) is 2:1, and the E/C ratio (electrolyte to the cathode capacity) is 2.5 g Ah⁻¹, which is a lean electrolyte condition.² The five points on this 2D colormap

represent well-matured technology (Cu/Al = 8/12 μm), state-of-the-art technology (Cu/Al = 6/10 μm), two future possibilities (Cu/Al = 4/8 μm and 3/6 μm), and the theoretical limit (Cu/Al = 0/0 μm). From the plot, the specific energy is clearly more sensitive to the thickness of Cu than Al, given that Cu is much denser than Al (8.9 versus 2.7 g cm⁻³). Moreover, the two future possibilities can increase specific energy from 370 Wh kg⁻¹ in state-of-the-art current collectors to 384 and 394 Wh kg⁻¹, representing increases of 3.8% and 6.5%, respectively. Such an improvement is already significant because the annual increase in Li-ion battery's specific energy is only 3%–5% nowadays. If the substrate could be completely removed, the specific energy would reach 425 Wh kg⁻¹ (increase of 15% over existing state-of-the-art). Calculation details are in section 1 of the [supplemental information](#).

The effects of current collector weight reduction is more significant for Li/S batteries than Li/high Ni-oxide batteries, given that the weight of electrodes and electrolytes is typically less. The Li/S system is less matured, meaning

that there are greater uncertainties in the mass of each component. Therefore, we plot the dependence of specific energy on sulfur loading, utilization, and E/S (electrolyte to sulfur capacity) ratios in [Figures 2B and 2C](#). Using a conservative estimate of E/S ratio (4) and utilization (60%), the specific energies at 5 mg cm⁻² loading are 262, 274, and 292 Wh kg⁻¹ for state-of-the-art technology (Cu/Al = 6/10 μm), a future technology (Cu/Al = 3/6 μm), and the theoretical limit (Cu/Al = 0/0 μm), respectively. Alternatively, for an optimistic estimation of E/S ratio (2) and utilization (80%), the specific energies at 5 mg cm⁻² are 476, 507 (6.5%), and 549 (15.3%) Wh kg⁻¹ for Cu/Al = 6/10 μm, 3/6 μm, and 0/0 μm, respectively. These values are higher than a previous analysis, such as 242 Wh kg⁻¹ at E/S ratio of 4, 60% utilization and 5 mg cm⁻², given that 10 μm-thick Cu foil and single side coating were considered there.¹ Calculation details are in section 2 of the [supplemental information](#). Analysis on other E/S ratios and sulfur utilizations can be found in [Figure S1](#).

These available increases in specific energy would more easily allow for long-sought-after goals without requiring exceedingly challenging sulfur loadings. For example, to reach 500 Wh kg⁻¹, 6.8 mg cm⁻² sulfur is needed for Cu/Al of 6/10 μm at E/S = 2 and 80% sulfur utilization. In contrast, only 4.5 mg cm⁻² is necessary for Cu/Al of 3/6 μm, which has been reported in literature. Moreover, only 1.9 mg cm⁻² is needed when the current collectors can be completely removed, which is readily achievable. The possibility of thinner current collectors should be considered in analyzing specific energy of next-generation technologies.

In the analysis above, the full removal of current collectors led to the largest gain in specific energy. Theoretically, the Cu current collector can be completely

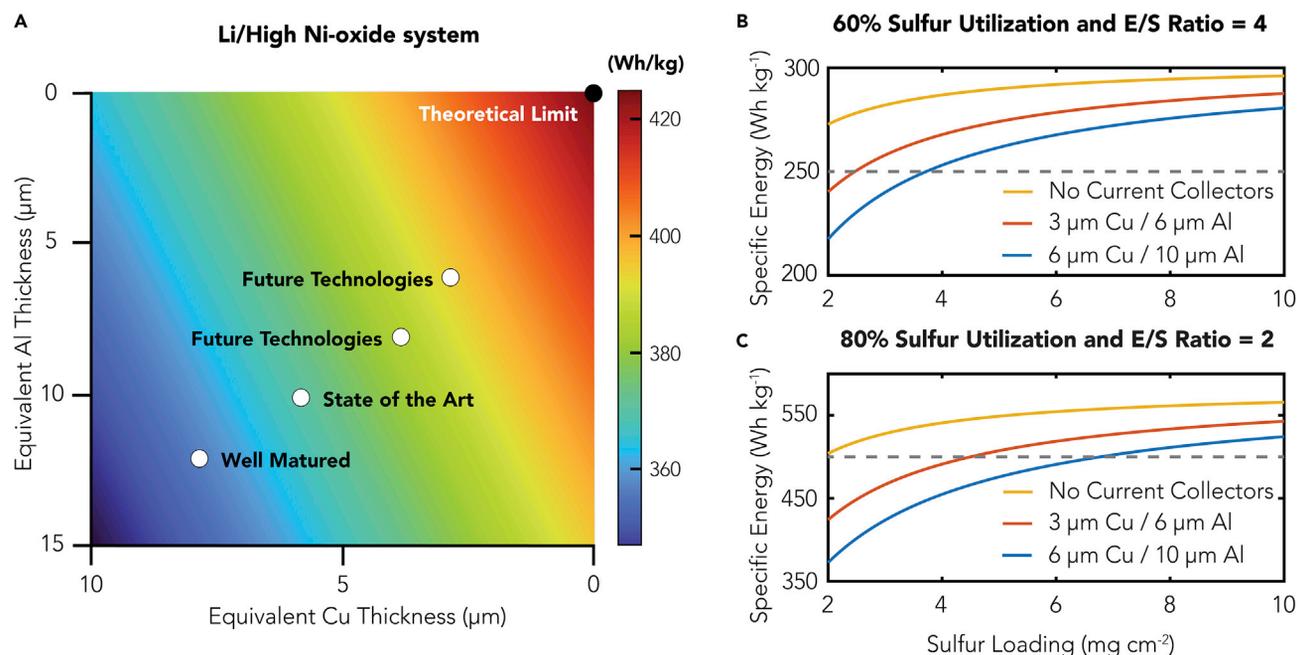


Figure 2. Impact of current collector thickness on specific energy

(A) A 2D colormap showing the benefit in specific energy (Wh kg^{-1}) by reducing the thickness of Cu and Al current collectors for a representative Li/high-Ni oxide cell with 3 mAh cm^{-2} (15 mg cm^{-2}) loading, an N/P ratio of 2:1, and an E/C ratio of 2.5 g Ah^{-1} . (B and C) Specific energy as a function of sulfur loading and current collector thicknesses are given for a Li-S cell with an anode to cathode ratio (N/P) of 3, (B) 60% Sulfur utilization and E/S ratio of $4 \mu\text{L mg}^{-1}$, and (C) 80% Sulfur utilization and E/S ratio of $2 \mu\text{L mg}^{-1}$.

removed because lithium metal also conducts electrons well ($1.1 \times 10^5 \text{ S cm}^{-1}$ for Li versus $5.96 \times 10^5 \text{ S cm}^{-1}$ for Cu). However, complete removal of the Cu substrate poses challenges such as surface inhomogeneities leading to non-uniform Li stripping and/or plating and a large current density localized near the tabs, resulting in significant increases of local temperature. Moreover, lithium becomes more granular during cycling, which significantly reduces electronic conductivity. This might be mitigated through the use of thicker Li, although the specific energy would be reduced appreciably (e.g., from 448 Wh kg^{-1} with $50 \mu\text{m}$ lithium to 371 Wh kg^{-1} at $100 \mu\text{m}$ lithium). Despite this, it should be noted that the Cu-free design has been explored in Li/S batteries.⁹ On the other side, it might be challenging to fully remove the Al substrate because the cathode materials have a low electrical conductivity of $\sim 1 \text{ S cm}^{-1}$, much lower than the desired value ($> 1,000 \text{ S cm}^{-1}$).⁸

METHODS TO PRODUCE LOW-WEIGHT CURRENT COLLECTORS

Currently, thin Cu foils are made by calendaring or electroplating, with electroplating becoming more dominant, although Al foils are generally made by calendaring. Developments toward lighter current collectors have also been explored through several varied strategies. Besides making thinner foils, porous foils and metal-on-plastic foils are two major strategies that are being actively explored.^{8,10–12} Takashishi et al. used lasers to create micro-holes on current collectors, which are typically $\leq 100 \mu\text{m}$ and account for $\leq 20\%$ of the original area (Figure 3B).¹⁰ Such holes not only help to reduce the weight of current collectors but also facilitate ion transport between the two sides of the current collector, benefiting balanced cycling and power density. One challenge of this strategy is that the slurry coating process becomes difficult because the slurry might fall through the holes before drying.

Moreover, a non-uniform perforation pattern could decrease the electrical conductivity. Dry coating methods might have the potential to address this issue. Chu and Tuan (2017) also reported a $1.2 \mu\text{m}$ thick, porous Cu foil by roll pressing Cu nanowires together (Figure 3A).¹¹

Elsewhere, the metal-on-plastic foil strategy can be realized by sputtering or evaporating metals onto a thin plastic foil (e.g., polyethylene terephthalate-PET), which is being explored in industry at the pilot level. The low density of plastic helps reduce the overall weight. For example, $0.5 \mu\text{m}$ -thick Cu on each side of a $6.5 \mu\text{m}$ -thick PET film is equivalent to the weight of a $2 \mu\text{m}$ -thick Cu foil, but with much better mechanical strength. Recent literature also shows that more functions can be incorporated by this multi-layered approach, such as using a fire-retardant polymer substrate to further enhance battery safety (Figure 3C).¹² Feedback from industry also

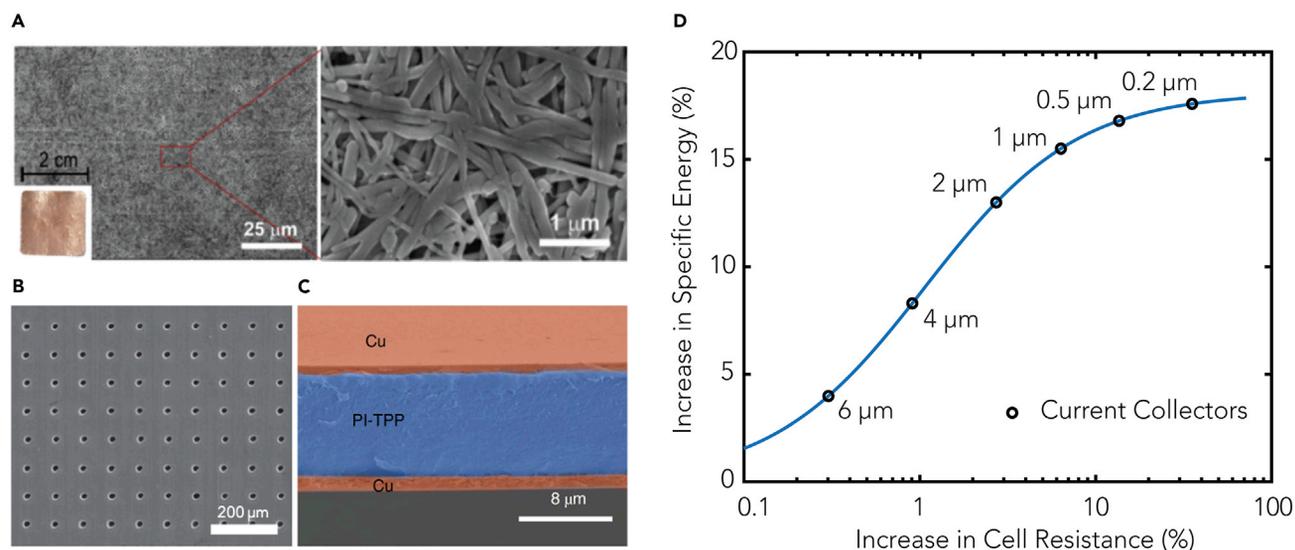


Figure 3. Recent work on current collectors and resistance effects

(A–C) Images of different low-weight current collectors.

(A) SEM images of a Cu nanowire foil current collector of varying thicknesses. Reprinted with permission from Elsevier.¹¹

(B) A porous Cu foil with 2.5% areal holes (diameter: 20 μm) diameter holes. Reprinted with permission from Electrochemical Society of Japan.¹⁰

(C) 500 nm Cu sputtered onto both sides of a polyimide-triphenyl phosphate (PI-TPP) supporting film. Reprinted with permission from Nature Publishing Group.¹²

(D) The increase in cell resistance and specific energy by using thinner current collectors, for a cell with the same parameters in Figure 2A. The plot assumes that both current collectors are of the same thickness, and the baseline thicknesses are 8 μm each. Calculation details are in section 4 of the supporting information.

suggests that this metal-on-plastic approach can reduce thermal runaway in nail penetration tests, possibly because of the electronic contact resistance between the current collector and nail increasing significantly, limiting the maximum current. However, it should be noted that the thicker metal-on-plastic film will decrease volumetric energy density slightly (e.g., 1%–4%), and a quantitative analysis is shown in Table S1. Such reduction in energy density should be analyzed for applications such as electric vehicles, but it would have less effect on aerial vehicle applications, where the volume is less important.

Finally, other materials altogether can be considered for future current collectors. For example, titanium is stable with the lithium metal anode and has good mechanical strength (tensile strength of 430 MPa versus 210 MPa for Cu). However, with inferior conductivity, processing, and cost, the application of Ti in vehicles might not be

currently feasible, but it could be considered for aerial and military applications, which might be less sensitive to cost.

POTENTIAL ISSUES OF LOWER WEIGHT CURRENT COLLECTORS

Although current collector weight reduction will certainly increase the specific energy of batteries, attention should also be paid to the challenges raised in other aspects. The first challenge to consider is the increase in cell resistance, given that the current collector resistance is inversely proportional to its thickness. For example, a typical 3 Ah cell with 20 repeating layers has an internal resistance of around 15 mΩ. A 6 μm Cu and 10 μm Al render an electronic resistance of 0.14 mΩ for the two current collectors together across all layers, but this electronic resistance quickly increases to 2.2 mΩ for 0.5 μm Cu and 0.5 μm Al, accounting for over 14% of the total cell resistance

as shown in Figure 3D. This effect will be larger for larger cells used in electric vehicles (e.g., 20–100 Ah). This challenge can be addressed by using multiple tabs so that the electronic current travels less distance on average. The thinner current collector will also reduce thermal conductivity along the foil direction, which is the in-plane direction in pouch cells and the longitudinal direction in cylindrical cells, given that heat is mainly carried by the metal foils in these directions. This will then reduce the cell's ability to dissipate heat, potentially increasing the likelihood of thermal runaway. This effect of thermal management and cell temperature also needs further studies (see section 5 in the supplemental information).

The second challenge is related to weaker mechanical properties in thinner films, which increases difficulties in electrode coating and calendaring, especially at high speeds

($\sim 1 \text{ m s}^{-1}$), as in current industrial manufacturing. As discussed above, this is less likely a problem with the metal-on-plastic strategy, given that PET's tensile strength is 170 MPa, comparable with Cu (210 MPa) and higher than Al (90 MPa). Finally, although lighter current collectors could decrease the raw materials cost of lithium-based batteries, more advanced processing techniques with increased costs remain as the main barrier to the effective integration of these developments to industrial manufacturing.

CONCLUSION

As electrode and electrolyte materials in high-specific-energy lithium metal batteries have become optimized, the percentage of "dead weight" has increased. These changes have made current collector weight reduction an attractive opportunity to increase the specific energy of cells by 10%–20%. Two representative lithium battery systems were analyzed to quantitatively understand the accessible gains through making current collectors lighter, demonstrating how, by optimizing one component, the requirements to meet long sought-after goals can be substantially eased in other areas of more challenging development. Multiple attempts at optimized and multi-functional current collectors have been recently developed. Although challenges remain, particularly with regards

to scalable production, the potential and incentives for improvement are clear, and advanced current collectors will be a key component in batteries for clean energy storage and numerous other future-oriented applications.

SUPPLEMENTAL INFORMATION

Supplemental Information can be found online at <https://doi.org/10.1016/j.joule.2021.03.027>.

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