



Commercializing Lithium Sulfur Batteries: Are We Doing the Right Research?

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A picture of the challenges faced by the lithium-sulfur technology and the activities pursued by the research community to solve them is synthesized based on 1992 scientific articles. It is shown that, against its own advice of adopting a balanced approach to development, the community has instead focused work on the cathode. To help direct future work, key areas of neglected research are highlighted, including cell operation studies, modelling, anode, electrolyte and production methods, as well as development goals for real world target applications such as high altitude unmanned aerial vehicles.

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Lithium Sulfur (Li-S) batteries are one of the most promising next generation battery technologies¹ due to their high theoretical energy density, low materials cost, and relative safety.² Li-S has the potential to achieve significantly higher gravimetric energy density than intercalation based lithium ion technologies,³ with some companies already reporting 400 Wh/kg cells.^{4,5} However, Li-S has a lower comparable volumetric energy,⁶ suggesting that applications where minimising mass is more important than volume will adopt it faster. Li-S technology is close to industrial production,⁷ with a number of companies scaling up manufacturing capabilities for large capacity cells.^{4,5} Meanwhile, the number of Li-S research papers published per year has increased dramatically from less than 50 in 2010 to over 900 in 2016. We have reviewed almost two thousand articles to identify the major gaps in research and discussed how targeting them could speed up the development and adoption of Li-S technology. We also discuss how from an industry/applied research viewpoint focussing on a performance metric, such as power density, would speed up development iterations, getting products to market sooner and help unlock further research funding.

Current Status

Li-S cells are already commercially viable in niche applications. In order to expand their market potential, however, there are still many challenges to overcome, such as limited cycle life, high self-discharge rates and over-heating at end of charge. Many of these are thought to be caused by the shuttle, where cathode species diffuse to the anode and react directly with the metallic lithium.⁸ Multiple solutions have therefore been proposed to prevent shuttle, such as physically^{9,10} or chemically¹¹⁻¹³ encapsulating the sulfur, designing tailored carbon structures,¹⁴ using electrolyte additives,^{15,16} separators,¹⁷ protective layers,¹⁸ or solid electrolytes to physically protect the anode.¹⁹⁻²¹ However, many of these solutions affect energy or power density adversely or do not function in practical commercial cells. Li-S batteries also undergo significant volume changes during operation, which poses a particular challenge for battery pack system designers and is being studied only since recently.²² These observations have only been possible since large form factor pouch cells are available. The effect of precipitation on useable capacity and reversible capacity loss,²³ and the influence of precipitation kinetics on rate capability,^{24,25} have also only recently been understood, after being observed in large cells. While rate limiting in commercial-size cells, these effects are probably never rate limiting for the high electrolyte to sulfur

ratios found in coin cells.²⁶ This highlights the importance of making practical and representative test cells and pre-commercial cells, based upon scalable manufacturing processes, as described by various articles.^{6,27-30}

To identify the key areas of current Li-S research, 1992 articles were sourced from Google Scholar using the search term “lithium sulfur”. Articles 1-999 had no date constraint, and articles 1000-1992 were limited to 2015-present (May 2017). Of the 1992 articles sampled, 1524 were relevant to Li-S, and were categorized by research topic, as described in their abstracts. One article could cover multiple topics. Articles reviewed included journals, conference presentations and posters. Figure 1 shows the distribution of topics addressed, in detail on the left and at a high level on the right. At this level of analysis, the results are similar to those reported previously.³¹

60 review articles examined overall Li-S research and suggested areas for investigation. The direction for research recommended by the most recent 34, covering all review articles published between 2014 and 2017, was synthesized in Figure 1 as ‘recommended areas’.

Figure 1 shows the research community has a strong preference for investigating novel materials design for the cathode,³² for the purpose of solving main issues in Li-S batteries, such as the low electric conductivity of sulfur and the polysulfide shuttle. Significantly fewer studies aim to remove these limitations by the design of other cell components, and even fewer are focused on non-material related aspects such cell operation and control. Contrary to the current approach, Figure 1 demonstrates that the review papers suggest a holistic, balanced approach to solving the problems faced by Li-S.^{10,27,33,34} The results of this review indicate the research community has largely ignored its own advice.

Future Needs and Prospects

Neglected research areas.—The Silicon Valley community regularly cites a paper³⁵ from 1943 during WW2 to exemplify the advantages of thinking differently about a problem. Bullet holes in aircraft returning from war were mapped and combined to identify where most aircraft had been shot. However, extra armour was not fitted to where the most bullet holes were observed, because these aircraft had returned. Instead the armour was fitted to where few bullet holes had been observed, because those aircraft usually did not return. In a similar fashion, it could be argued that directing efforts toward neglected rather than well studied areas of research increases the potential for significant breakthroughs. As shown in Figure 1, the research community appears to know already which these areas are, but there has been limited reallocation of time and effort. Here we look further into each area and discuss its potential.

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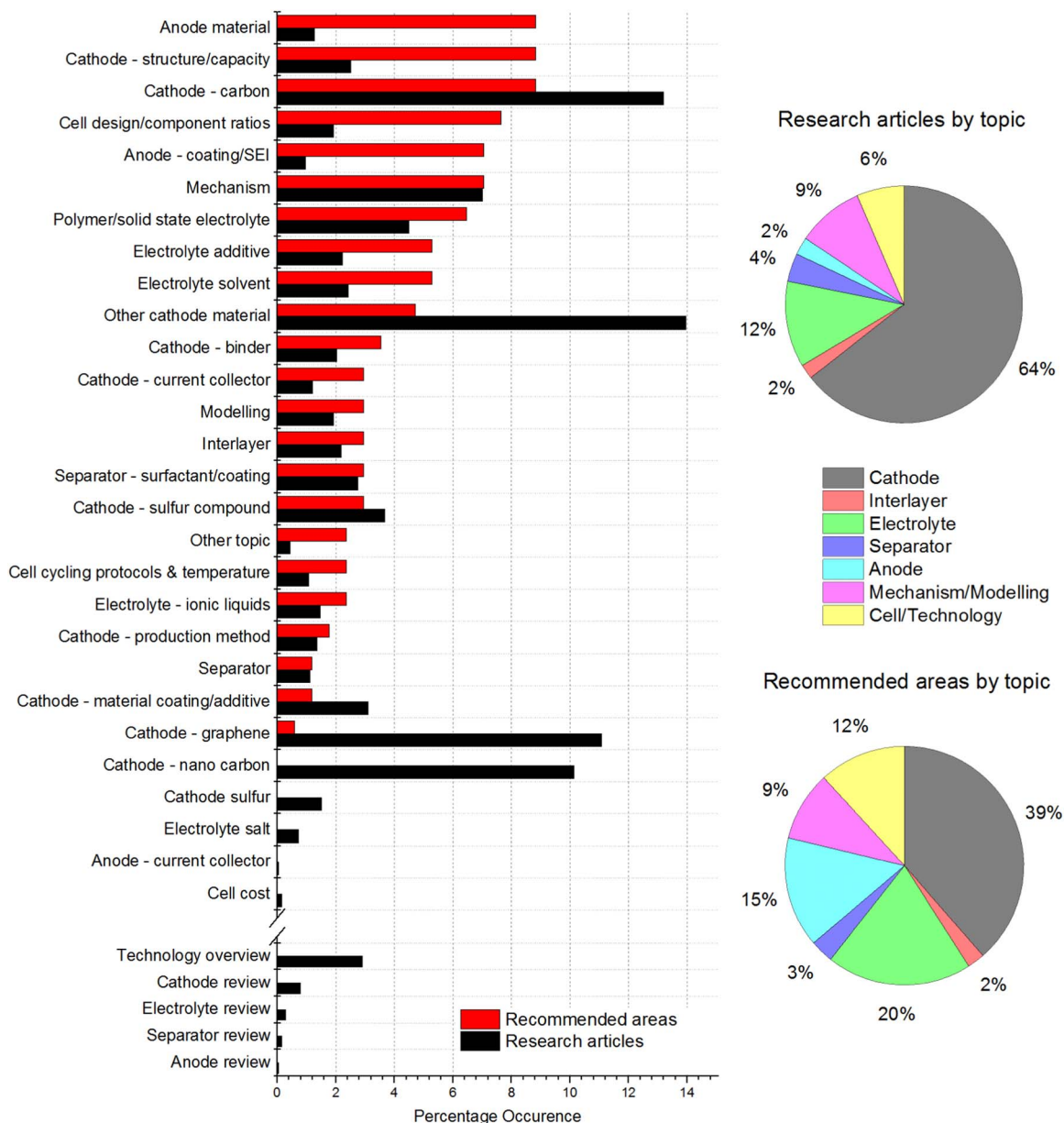


Figure 1. Comparison of topics addressed in research articles, from the abstracts of 1524 articles, versus recommended topic split that has been defined in review articles.

Cell operation.—Cell performance in real applications can differ significantly from that in the lab, as a result of ‘non-ideal’ current loads, including incomplete charge/discharge cycles, noisy current loads and rest periods, but also thermal and electric connections inherent to a battery pack. Only 14 articles, however, follow the effects of operational parameters on cycling performance, and only two the effects of storage or idle time. Equally surprising, 7 articles analyze the effect of discharge rate on cycling, and only two that of charging rate, despite widespread awareness³⁶ that charging parameters affect both cell performance and capacity fade. Only 4 consider the effects of temperature.

The widely reported and strong dependence of cell capacity on charge/discharge rates and temperature is partly due to accumulation of precipitated Li_2S ³⁷ and shuttle,³⁸ the two key mechanisms determining the capacity fade of the cell. Probably a consequence of these mechanisms, a significant history effect has been demonstrated in Li-S.³⁶ This study has crucial implications on data reproducibility, validity of material choice, and tailoring of operational parameters.

Therefore, the history effect must be thoroughly understood and quantified, yet it has received little to no further attention.

Bespoke testing, together with further development of mechanistic models and computer-driven data analysis are crucial to understanding history effect and to successfully choosing operational parameters for optimum charging and cycling of the cell.

Control modelling.—Equally important to Li-S deployment is the availability of tools for state-of-charge and state-of-health estimation. Because of its unique features, such as the shuttle, Li-ion control techniques are not applicable to Li-S.³⁹ There are only 4 articles that model Li-S for control, published by two collaborating groups.^{40,41} Additional effort is clearly required in this area with essential impact on the future of Li-S.

No successful attempts of SoH estimation for Li-S batteries are published. Estimating SoH requires novel diagnostic techniques that can track degradation, such as the Differential Thermal Voltammetry technique for Li-ion batteries,^{42,43} as well as a good mechanistic

understanding of Li-S degradation, obtained from physical modelling and characterization. Current degradation models consider only loss of sulfur species during shuttling and irreversible precipitation,^{38,44,45} ignoring solid-electrolyte interphase (SEI) growth and electrolyte depletion.⁴⁶

Anode.—Almost all review papers mention the anode as a critical area for development, mainly to suppress the polysulfide shuttle, yet only 46 articles studied the anode, compared with 1273 that studied the cathode. The majority focused on additives, with 24 explicitly studying LiNO₃. However, cells containing LiNO₃ produce gases and swell above ~40°C,⁴⁷ rendering them impractical because they cannot pass Test 2 of UN38.3 Transport of Dangerous Goods Certification.⁴⁸ The effective use of additives and the formation of a stable SEI is made more challenging by the fact that the stripping and plating of the lithium anode results in significant changes in thickness,²² estimated to be around 10 μm for each 2 mAh cm⁻² of cathode surface capacity.⁴⁹

Despite the fact that the separator plays a significantly more important role in Li-S than in Li-ion, 57 articles looked at coating off-the-shelf Li-ion separators, and only 23 at developing a bespoke separator for Li-S. Hassoun and Scrosati proposed moving to a solid-state configuration back in 2010¹⁹, and 93 of the articles have studied solid-state or polymer electrolytes. However, very few articles looked at creating a layer on the anode and using a conventional liquid electrolyte,^{50,51} which is probably a more promising near-term path to a commercial cell with close to zero shuttle, assuming such a coating is cheap and can be mass-produced. From the studies that used conventional electrolytes, of the 20 that mentioned coatings only 4 developed an ex-situ, non-additive based coating.

Electrolyte.—As mentioned, 93 articles have investigated polymer and/or solid-state electrolytes; there were also 36 articles on ethers, 30 on ionic liquids, 16 on dioxolane, 7 on carbonates, 5 on sulfones and 13 on “others”. This highlights a trend to look at too few solvents.¹² More primary solvents and solvent combinations, as well as lithium salts and additives, should be explored to optimize performance, due to the major role that the electrolyte plays within the cell. Electrolyte research also must target significantly increased operational temperature window (<0°C or >30°C), because low or high temperature performance is essential for mainstream applications and for most battery certifications.

However, the biggest neglect appears to be translating results from coin cells with very high and unrepresentative electrolyte to sulfur (E/S) ratios (>10 μL mg⁻¹) and low areal sulfur loadings (<2 mg cm⁻²) to those necessary for high energy density pouch cells (3–4 μL mg⁻¹ and >8 mg cm⁻²).²⁶ Low E/S ratios define the cell's performance in practical cells,²⁷ through mechanisms such as polysulfide solubility, electrolyte viscosity, S/Li₂S precipitation kinetics, and cell lifetime. Different E/S ratios can lead to a different mechanism being limited, such that, unfortunately, many published results are not transferable to practical devices.

Production.—While 40 articles mention cell design and 28 discuss cathode production, most other work does not follow the suggested guidelines regarding realistic cell design and scalable production methods. For the production of Li-S cells in large quantities, the manufacturing processes must be fast, to keep costs low. For example, to produce 300 million batteries a year (3.4 GWh, enough for 50,000 cars), as is the case for Li-ion Panasonic 18650⁶, the coating throughput must be 348 m per minute. Therefore, coating methods of at least 50m per minute are typically needed to ensure the cost of the coating machines is not prohibitive.

To achieve costs below \$100 kWh⁻¹, all the materials within the cell must be cheap, abundant, and produced at a large scale. For example, the scale of materials required for current Li-ion cathode manufacturing is on the order of 10 000 tons per year⁵² and will likely rise to the order of 100 000 tons per year. These numbers indicate that some currently proposed material solutions are not applicable in mass produced Li-S batteries, and that materials making use of existing

supply chains are more likely to succeed. Materials and production methods should also be non-toxic to reduce the environmental impact, restrictions and cost. A number of common solvents, such as NMP, DMF, or DMAc, have become regulated by EU's REACH and are thus quickly being substituted in many industries.⁵³ Therefore, more focus on green solvent alternatives is needed.

Development path.—The progress of the Li-S development could be greatly accelerated by using a rapid research and test method, setting suitable goals and defining a clear path to achieve them.

Accelerated testing and development.—Picking the right development path will help cut cost and time to market - for example, by developing power capability of a cell first, one can cut overall development time and cost because cycling is faster, delivering results faster, which in turn allows research decisions to be made faster, as shown in Figure 2. This method is akin to the Fail Fast development methodology used in other sectors.^{54,55}

Research^{56,57} has indicated that the traditional techniques used to accelerate Li-ion life testing, such as high power combined with high temperature cycling can deliver misleading results, leading to expensive warranty claims and reputation damage for companies concerned.⁵⁸ For Li-S, this problem could be exacerbated, as the mechanism is more complex. Therefore new, accurate, testing techniques and performance models need to be developed for Li-S to ensure that SoH can be predicted reliably, allowing batteries to be warranted and deliver performance.

Accelerated development such as this is critical to companies trying to commercialize Li-S, as substantial investment is required to bring a product to market, so any time savings will also save cost. This is also applicable to publicly funded research groups, as funding bodies are more likely to fund follow on work when larger quantities of high quality results are reported. The development path is quickest when a realistic application is targeted by the development teams, guiding decision making throughout the process.

Target applications.—Li-S cells are most likely to enter the markets where mass is the critical factor above all else. Persistent unmanned aerial vehicles (UAVs) such as the Airbus Zephyr,⁵⁹ Project Loon⁶⁰ and Facebook Aquila,⁶¹ which slowly charge their batteries during the day and discharge them overnight, are likely candidates for the initial adoption of Li-S in the near future. Following that, a number of concurrent development paths would target the wider aerospace and other adjacent markets, such as space⁶² and automotive, for which better power and cycle life is required. For automotive, Li-S would be suitable for applications where payload is critical but volume is less important, such as buses and trucks, rather than consumer vehicles.

The timescales associated with these developments are difficult to predict, as they depend on both the speed of development and passing essential regulatory certifications for each market, such as automotive,⁶³ aircraft⁶⁴ and satellites.⁶⁵ Therefore, although limited adoption of Li-S is expected in the near future, it will probably take a number of years for Li-S to become as widely available as Li-ion cells are today.

Conclusions

To date, the research community has mainly worked on the cathode rather than take a balanced approach. We recommend that future development should take a holistic, comprehensive approach and focus on these key points:

1. Develop and use realistic test cell designs, balancing the component ratios to deliver accurate performance of the materials.
2. Appropriate research prioritization, that follows the advice of the various review papers to increase work in particular areas currently neglected.

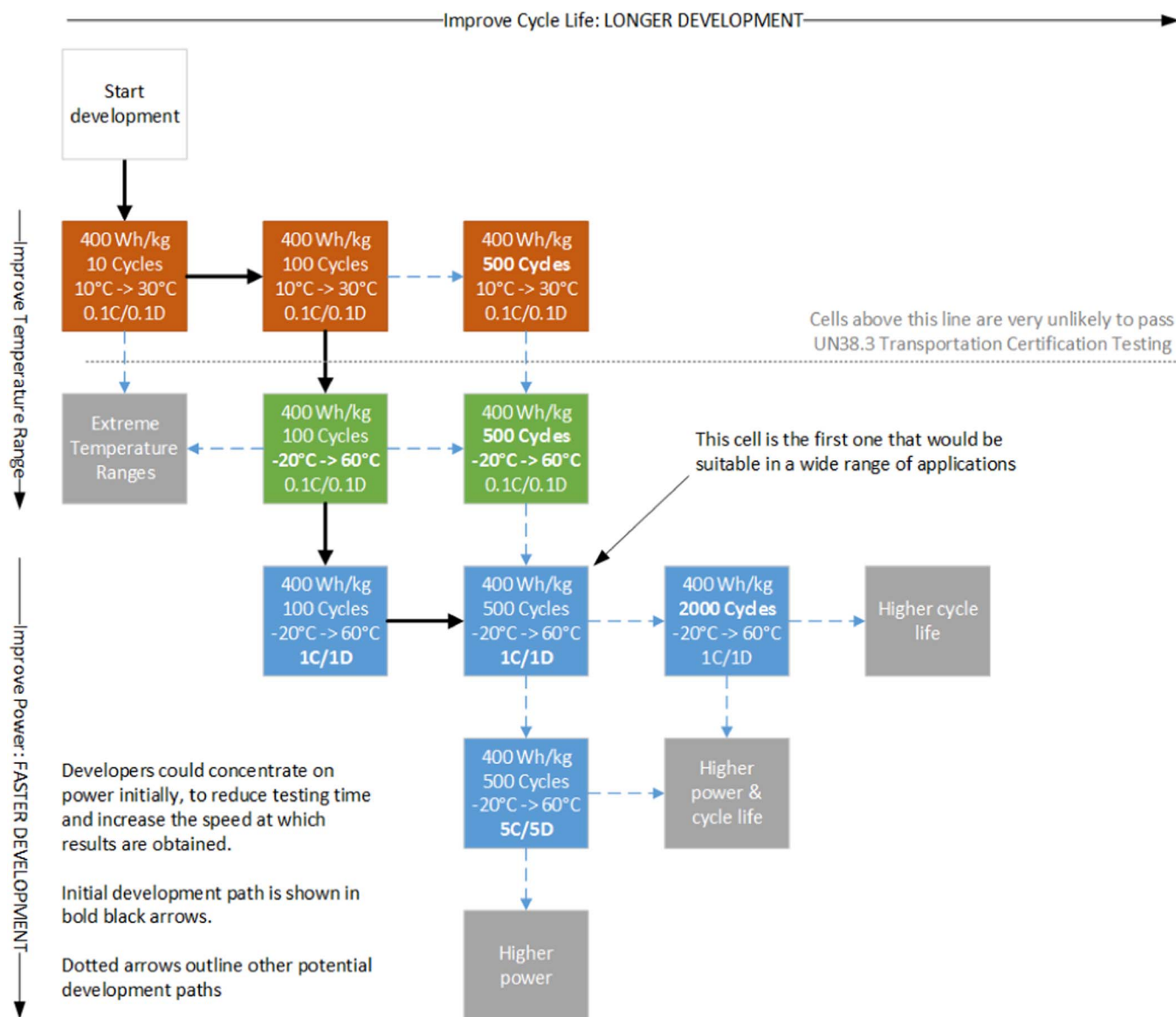


Figure 2. Example accelerated development path focused on power to reduce time to market.

3. Develop a greater understanding of the Li-S mechanism, not only to improve materials research, but also to build mathematical models to allow the cells to be controlled in real applications.


In addition, for development aiming to use Li-S cells in real world applications, we recommend the following:

1. Targeted cell design and performance for a suitable application, such as a persistent UAV, where a high specific energy, charge/discharge rates of 0.1C/0.1D⁵⁹⁻⁶¹ and a cycle life of 200⁶⁰ is acceptable. Longer term, improved cycle life and power will be required to make Li-S suitable for other applications.
2. Use of viable mass production methods that can easily be scaled up to the levels required for commercial sale.
3. Development of chemistries that comply with regulations such as UN38.3 so that the products can be shipped and sold.
4. Focus on performance metrics that speed up testing, such as power density, enabling design iterations to occur faster, leading to earlier market entry, reduced development cost and potential public research funding increases.

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