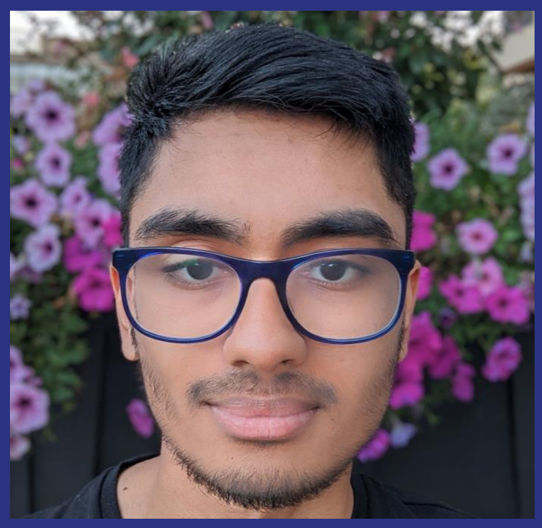
Improving Zero-Excess Li_2S cells through optimisation of electrode coating

Preparation Li_2S cathodes through doctor blading to improve scalability



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Abstract and Introduction

- Lithium sulfur (**Li-S**) Zero-excess-lithium (**ZEL**) batteries have the potential to become the next generation of batteries for electrical vehicles and portables, owing to their **greater energy density** and **lower production costs**[1].
- However, many challenges are still faced as research into this cell configuration is scarce, mainly due to **shuttling** and **volumetric changes**[2].
- One approach presented here is to alter the proportion of our cathode composition, containing lithium sulfide, carbon, binder, and solvent.
- **Coin cells** containing cathodes of these compositions are created through **doctor blading** the produced slurry onto an aluminum carbon foil for **electrochemical testing**; the results discussed on this poster.

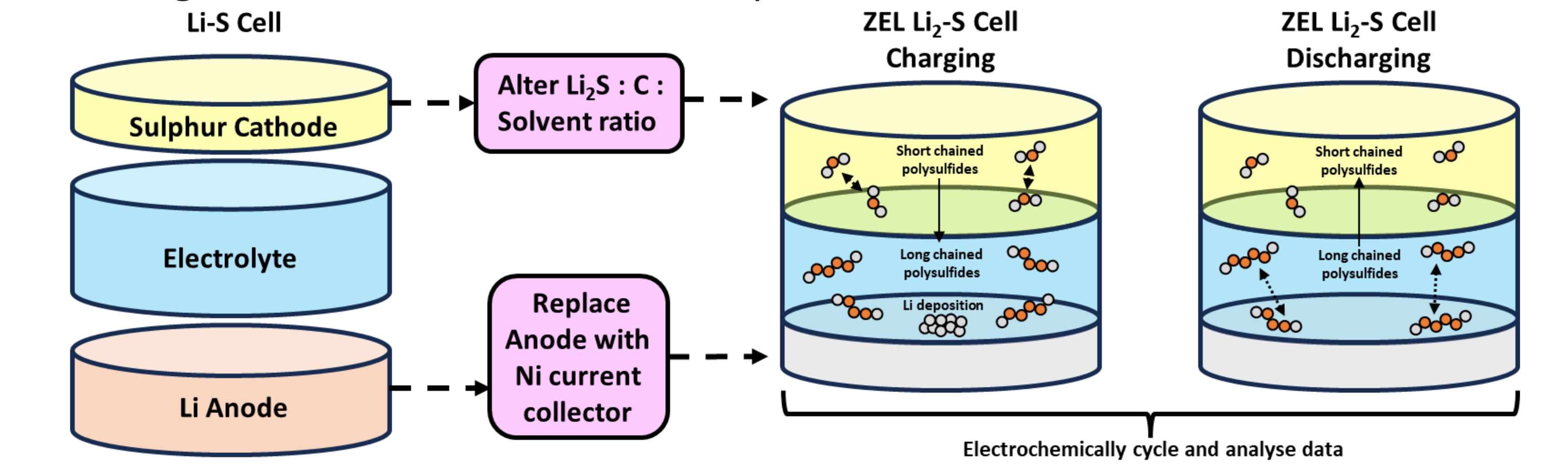


Figure 1. Schematic diagram showing Li-S Cell, ZEL Li-S cell and polysulfide formation during charge and discharge.

Motivation

- **Li-S** batteries offer a **high theoretical specific capacity** (1675 mAh g^{-1}) while maintaining a **low per unit cost** due to the abundance of sulfur[3].
- However, they still face **safety issues** due to dendrite formation from the Li anode, as well as **poor reversibility** and **columbic efficiency**[4].
- These could be mitigated with the proposed **ZEL** cell configuration, replacing the Li anode with a nickel current collector, allowing for a much **greater energy density** owing to the **volume and weight reduction**, and **increased safety** due to limited Li.

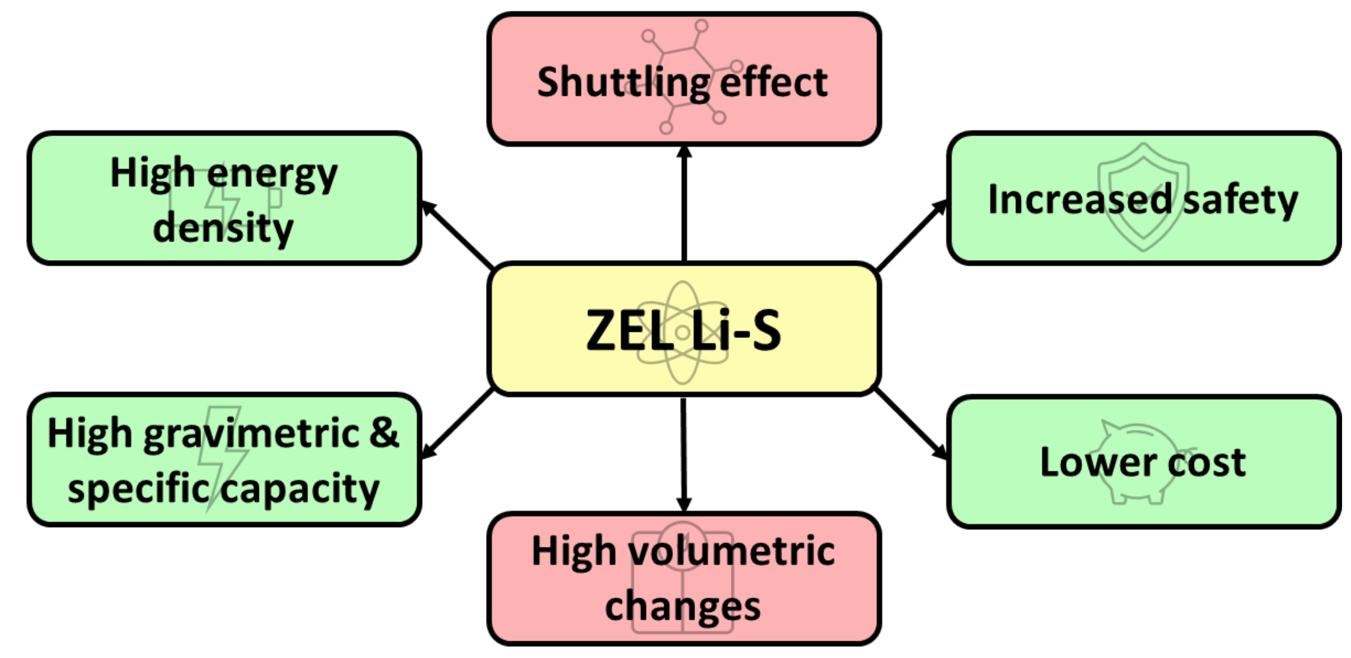


Figure 2. Spider diagram of the advantages and disadvantages of ZEL Li-S cells compared to Li-S cells.

Method

- Powdered Li_2S + carbon, and 10% binder solution (SBR or PVDF) + solvent (toluene or NMP) were weighed and poured into pot within a glove box. Mass weighed was calculated using densities, dependent on ratio of compostion wanted (for example 40:45:10 wt.% Li_2S:C:PVDF). Slurry was placed inside a mixer for 20 minutes at 2000rpm.
- Slurry was doctor bladed with an 80mm blade gap carefully onto an aluminium carbon conductive foil, and left to dry overnight at 50°C.
- Cathodes were crimped out and constructed into coin cells for electrochemical cycling for an activation cycle at C20, and then 100 cycles at 1C. Method repeated for different ratios.

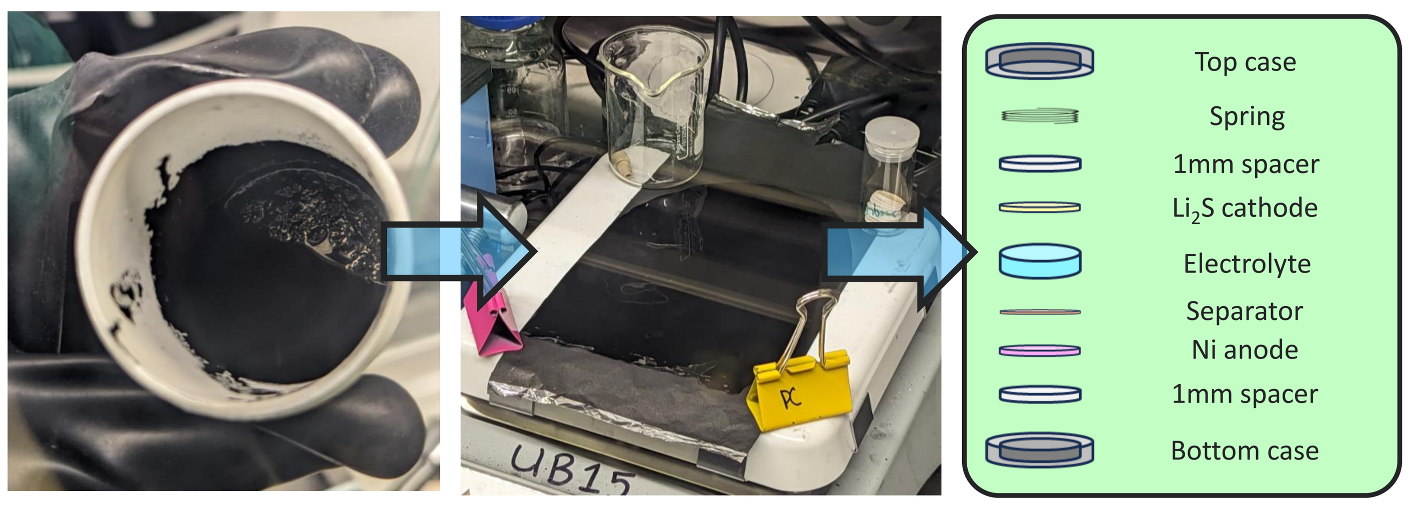


Figure 3. Images of a slurry, and after doctor blading onto foil onto a hot plate. Schematic of coin cell construction.

Results

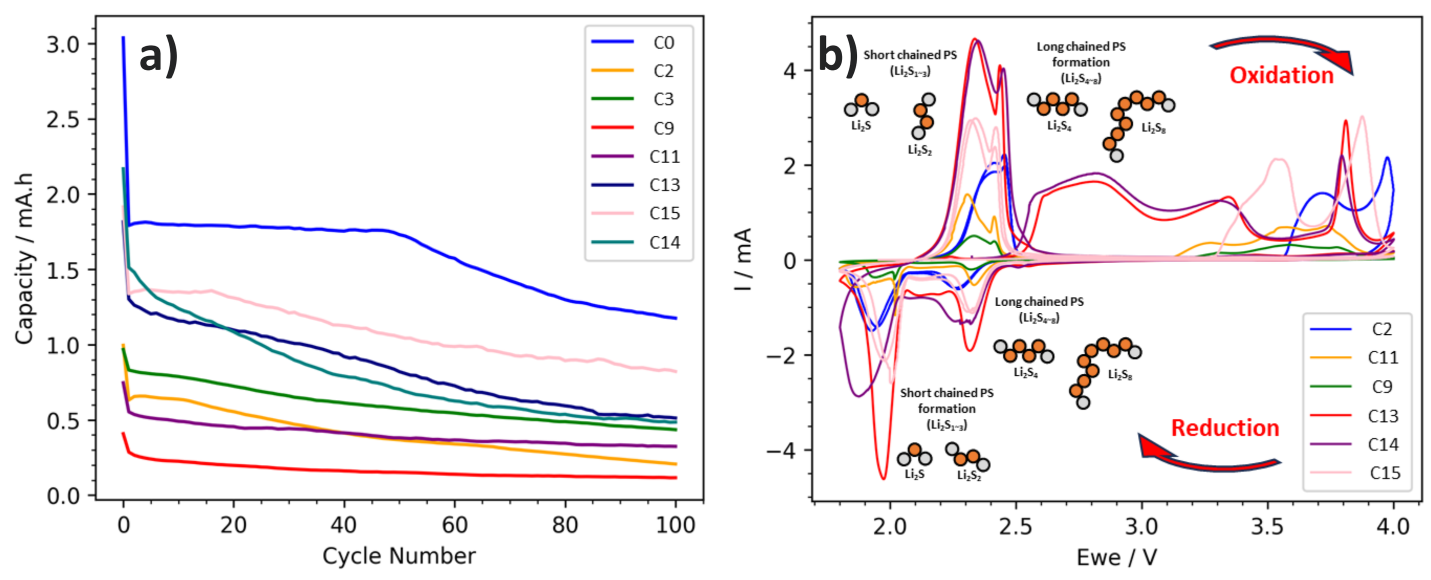
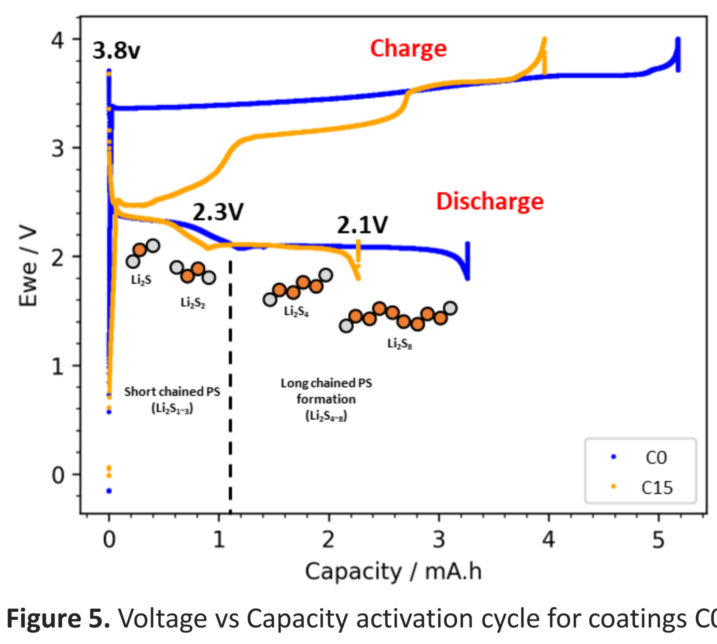


Figure 4 a) Capacity vs Cycle number for all coatings shown in Table 1, C0 showing the highest capacity with fade after ~50 cycles, and C15 following. **b)** Cyclic Voltammetry curves for a few coatings, with anodic/cathodic illustrations.



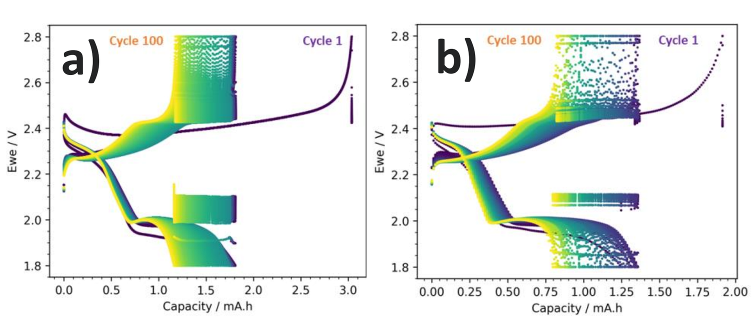


Figure 6 Voltage vs Capacity after 100 cycles for **a)** C0. **b)** C15.

	C0	C2	C3	C9	C11	C13	C14	C15
Li_2S / %	70	50	45	40	30	40	50	60
C / %	20	35	20	50	55	45	35	25
SBR / %	10	15	35	-	-	-	-	-
PVDF / %	-	-	-	10	15	15	15	15

Table 1. Table of all coatings and compostion ratios created.

Figure 5. Voltage vs Capacity activation cycle for coatings C0 and C15, with redox reactions illustrated.
All graphs were produced using a self-written GUI application coded in Python given .txt data (code on **GitHub**)

Discussion

- Capacity and quality of coating improved as the Carbon content was decreased from 55% to 25%, and Li_2S increased from 30 to 60% when using PVDF binder + NMP solvent; the best being C15 (**60:25:0:15** with a **discharge capacity of ~0.8 mA.h** after 100 cycles). Comparably this is lower than the standard C0 coating using SBR + toluene (**70:20:10:0** with a **discharge capacity of ~1.2 mA.h** after 100 cycles). A coating of **70:15:0:15** was also created, however quality was too poor as it would not stick to foil, suggesting that the binder solution should be increased from 10%. Many coatings of differing ratios were created (C1-16), however due to poor coating quality no cathodes could be made. Most cells shown here had **distinct peaks** on their CVs and plateaus on their activation cycles, corresponding to **long and short chain polysulfide formation** and redox process' at 2.1V and 2.3V, matching that of literature.

Conclusions

- Cells created with **PVDF binder + NMP** solvent **performed better electrochemically** in general compared to SBR + toluene, with **improved quality of coating.**
- While the standard C0 coating performed the best, there is a ratio between **60:25:0:15** (C15) and **70:15:0:15** which may perform better than C0.
- **Minimizing the conductive carbon** while **maximizing the active component** Li_2S shows **heightened performance**.
- Slurry should be **dried slowly** to ensure **no quick volumetric changes** and **cracking**.

Impact / Next steps

- There is an identifiable correlation between **carbon : Li_2S ratio and capacity**, and therefore more composition ratios should be tested for optimal electrochemical performance.
- **EIS** and **SEM** should be used for better understanding into the morphology of cathodes, specifically pores and surface area.
- **Alternative** binders, solvents, and conductive foils should be experimented.
- Cells should be **cycled at varying rates**.

References

- [1] - A. Bhargav, J. He, A. Gupta, A. Manthiram, Joule 2020, 4, 285.
- [2] - L. Zhou, W. Zhang, Y. Wang, S. Liang, Y. Gan, H. Huang, J. Zhang, Y. Xia, C. Liang, Journal of Chemistry 2020, 2020, 1.
- [3] - R. Mori, J Solid State Electrochem 2023, 27, 813.
- [4] - S. Nanda, A. Gupta, A. Manthiram, Adv. Energy Mater. 2018, 8, 1801556.

Intern bio

- Navraj Eari is entering his 4th year studying MEng Materials science and Engineering at Imperial College London.
- Keen developing his knowledge in computation and battery storage technology.
- Hoping to undertake a masters project and future career post graduation in related field.
- Outside studies, he enjoys playing video games and basketball.

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