Background

Current state-of-the-art commercial LIBs are based on a battery architecture that uses a liquid structure (electrolyte) as the electric insulation layer between the electrodes. The most challenging problem in such electrolytes is dendrite formation: growth of nano-sized tree-like lithium structures on the anode and into the electrolyte. These dendrites conduct electricity, defeat the electrolyte’s purpose, and pose the risk of battery explosion.

Solid-state LIBs are considered the solution for these drawbacks of liquid electrolytes. In solid-state LIBs, the electrolyte is solid rather than a liquid (**solid-state electrolyte**, SSEL), and therefore is less susceptible to dendrite formation (*but do not eliminate them completely*), less sensitive to temperature changes, and less susceptible to explosion. Importantly, the reported energy densities of SSELs are capable of satisfying the energy needs of electric vehicles while ensuring lower battery volume and longer driving range.1

Theoreticians and experimentalists alike have been able to discover a large number of materials as candidates for solid-state LIBs. Theoreticians have benefitted from the near-negligible cost of material exploration in comparison to the costs borne by experimentalists when they synthesise new battery materials. The principal theoretical approach for modelling SSEL materials – density functional theory (DFT), has been particularly successful in accurately predicting key SSEL parameters such as the electronic bandgap and the diffusion coefficients.2 Compounded with high-throughput screening workflows3,4 and applying machine learning,3 thousands of candidate materials have been examined, from which new materials have been proposed**. However, these materials have not yet been commercialised due to several shortcomings: electrode/electrolyte interface problems, dendrite formation and mechanical instability.**

Challenges

Presently, solid-state LIBs are based on **bulk SSEL** materials (such as the popular superionic conductor Li10GeP2S12). In spite of the successful incorporation of bulk SSELs into batteries (the solid-state LIBs), there are many challenges which have delayed their commercialisation:

1. *The electrode/electrolyte interface problem:* Complex electrochemical phenomena at the interface give rise to the structure known as solid-electrolyte interphase (SEI) which causes capacity fade of the battery.
2. *Lithium dendrite formation:* The growth of lithium dendrites at the anode/electrolyte interface is one of the key challenges in today’s commercial LIBs. Solid-state electrolytes have the ability to impede the formation of lithium dendrites. However, in some situations, lithium dendrites still form in the solid-state electrolyte.
3. *Mechanical stability:* Many of the known SSEL materials such as Li10GeP2S12 cannot sustain mechanical duress, which speeds up their deterioration.
4. *Lack of modular material design:* The thousands of materials that have been theoretically examined have been routinely harvested from existing online databases, such as Materials Project.5

2D battery materials are ideal candidates for solid-state LIBs, and are currently expanding into many avenues in lithium-ion battery research and many achievements have been experimentally realised.6,7 **Importantly,** **they directly address the above challenges:**

1. *They reduce interface resistance* because of the easier diffusivity of lithium from the anode/cathode into the gap between the 2D layers.6
2. *They are more able to suppress dendrite growth* owing to synergistic effects that lead to uniform distribution of Li ions between the 2D layers.6 Artificial solid-electrolyte interphase (ASEIs) based on 2D materials have been reported to reduce the likelihood of dendrite formation at the anode.8
3. *Highly mechanical stable materials can be synthesised* by blending different 2D materials with high hardness.9
4. *They allow for modular design*, where one can stack different types of materials to generate new materials, owing to the weakness of the bonding force that holds the layers together (the van der Waals interaction). Given that there are > 6,000 2D materials currently known, the number of possible pairs is more than 20 million. LEGO-like assembly of 2D materials into hybrid van der Waals heterostructures has been theorised in the Nature Perspective article by Geim and Grigorieva,10 and has drawn immense interest from experimentalists, who have successfully synthesised many hybrid van der Waals materials.

Proposal

Today’s solid-state lithium-ion battery architectures are based on a solid lithium anode (in principle the highest capacity anode material), a bulk (in contrast to layered) SSEL such as Li10GeP2S12, and a layered cathode such as LiCoO2. This battery architecture is represented as class 1 in Figure 1. This project aims at achieving the following conceptual and technical innovations to realise classes 2, 3 and 4 in Figure 1. The application innovations will offer spent battery materials a chance to be useful as catalysts for important chemical reactions.

* *Conceptual innovations:*
  + Hybrid van der Waals 2D materials have attracted immense attention recently for battery applications.9,11,12 Battery components based on hybrid van der Waals 2D materials will be selected for the electrolyte, cathode and anode components of solid-state lithium-ion batteries.
  + The single material architectures that this project will develop (class 4, the top class in Figure 1) are particularly innovative, and are inspired by the single material battery architecture that was introduced seven years ago,13 where the three battery components (anode, electrolyte, cathode) are made from the same material.
  + Inspired by the recent $50m funding to the company Amprius for manufacturing silicon nanowire anodes, layered silicon-lithium structures will be considered as candidate anodes for layered lithium-ion battery architectures.
* *Technical innovations:* 
  + Based on the ROSA descriptors14,15 and potential variants, machine learning models will be trained and new machine learning features will be generated to predict the key physical and chemical traits of battery materials to enable the rapid screening of hybrid van der Waals 2D materials for the construction of the ideal battery components for the architectures in classes 2, 3 and 4 in Figure 1.
  + The feasibility of the synthesis of these materials by **electrostatic layer-by-layer assembly**9,11 has been **established** by several recent reports of successful synthesis, such as Ti0.87O2/nitrogen-doped graphene,16 MnO2/graphene,17 VOPO4/graphene,18 MoS2/graphene.19 In order for the proposed battery materials to be feasible for experimental synthesis, this project proposes materials that can be synthesised by electrostatic layer-by-layer assembly.
  + The project extends my work in examining the structure and properties of spent batteries.20
* *Application innovation:* Solid-state LIBs have also lent themselves to after-life reuse in other applications including catalysis, supercapacitance, waste water treatment, and graphene synthesis.21 For example, a spent cathode was demonstrated to exhibit “bifunctionality” when it was found to catalyse both oxygen evolution and oxygen reduction reactions, and this was explained by DFT calculations.22 High-throughput screening procedures are currently being developed to examine the utility of a spent battery material in catalysis.

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