Tracing polysulphide speciation in salt-free semi-liquid Li-sulphur cells through in-situ Raman Spectroscopy

Matthew Sadd [a], Marco Agostini [a], Aleksandar Matic [a]
[a] *Chalmers University of Technology, Department of Physics, 41296 Göteborg, Sweden*

E-mail: sadd@chalmers.se

With the recent rise in the interest to develop Electric Vehicles (EVs), due to environmental concerns, one factor comes under more scrutiny than most – the ability to develop EVs with a range of over 500 km on a single charge. Additionally, we also see increased demand for our portable electronics to have longer lifetimes, as was most recently demonstrated at Consumer Electronics Show (CES) 2018. Lithium Sulphur (LiS) technology has the potential to meet these previously stated requirement as well as perform in large scale energy applications with a high theoretical specific capacity (1675 mAh/g) and a high energy density (2500 Wh/kg). In addition to boasting high theoretical values, the LiS technology carries the advantages of being a sustainable alternative to current Li-ion systems due to the abundance, low price and low toxicity of sulfur.

Despite the many advantages of a LiS systems, there are also many factors that currently hold it back from commericialisation: low practical capacity/energy density, short cycle-life and low rate capability. Many issues faced by the LiS battery systems stem from the parasitic 'shuttle' mechanism at the anode and cathode dissolution.

We have recently reported on concepts providing a route to overcome issues with practical capacity, rate capability and cycle life. One approach to increase the energy density used a graphene aerogel with high sulphur loading [1], this provided a self-standing cathode requiring neither binder and carbon black nor current collector. A further strategy was to use a 'catholyte' concept by dissolving the sulphur active material in a cell's electrolyte [2], demonstrating higher areal capacity.

Commonly, in a 'catholyte' cell, sulfur is dissolved in the electrolyte, which differs in approach to conventional LiS cells where polysulphides are formed and dissolve into the electrolyte during the electrochemical processes. In this scenario, development of techniques able to monitor the state of sulfur speciation are of high importance to accurately determine the processes occurring in the cell, thus allowing the improvement of a cell's life. In this contribution, we show how by employing the use of *in situ* Raman spectroscopy [3], the speciation of polysulphides at different states of charge/discharge are monitored to clarify the mechanisms of cells based on catholyte configuration in Li-salt free systems. This carries the advantages of removing fluorine-based species from within an electrochemical cell, and the possibility to further reduce the cost of LiS cells.

References:

[1] F. Nitze, M. Agostini, F. Lundin, A. Palmqvist and A. Matic, Scientific Reports 6, 39615 (2016).
[2] M. Agostini, S. Xiong, A. Matic, and J. Hassoun, Chem. Mat. 27, 4604 (2015).
[3] M. Agostini *et al.* ChemSusChem. 10, 3490 (2017).