Silicon-graphene anodes for long cycle life lithium ion batteries

D. Maxwell[1,2] A.R. O. Raji[2], J. Marcellino[2], A. C. Ferrari[2], C. P. Grey[1]
[1] *Department of Chemistry, University of Cambridge, Lensfield Road, Cambridge, CB2 1EW, UK*
[2] *Cambridge Graphene Centre, University of Cambridge, Cambridge, CB3 0FA, UK*
dcm44@cam.ac.uk

The development of high energy density Li ion batteries is essential if we are to succeed in the electrification of the transport system[1]. Central to this is the need for electrodes with increased specific capacity (mAh/g and mAh/cm^3) which can run for 100s to 1000s cycles[1]. The manufacturing of these electrodes must also be cheap and scalable in order to meet the demands of future electric vehicles. Here we focus on high capacity Si anodes. Si offers high theoretical capacity (3579 mAh/g)[2], nearly 10 times more than graphite[2]) and a low (0.1 - 0.4 V vs Li/Li^+) operating voltage[2]. However it undergoes up to 300% volume expansion upon reaching the fully lithiated $Li_{15}Si_4$ phase[2]. This causes both particle and electrode fracture[3], leading to continuous electrolyte decomposition at the electrode surface[3], forming a solid electrolyte interphase (SEI)[3]. This results in irreversible consumption of Li[4], impedance increase[4], and capacity fading[4]. Carbon coatings and additives can enhance cyclability[5,6], as well as electrical conductivity of Si electrodes[5-7]. Carbon coatings can also help manage the stress and strain of the electrode when Si is lithiated[5], reducing pulverisation[5]. These approaches are costly[6] and not compatible with commercial manufacturing techniques[1]. Here we show that graphene produced by microfluidization [7] can enhance the cyclability of Si anodes, more than doubling the capacity retention compared to more conventional Si composite anodes with carbon black[8]. Si-graphene composite electrodes are produced through a scalable, one-step mixing process. We combine a prelithiated Si-graphene anode with state-of-the-art nickel manganese cobalt oxide cathodes, resulting in >480 Wh/kg energy density and ~75% capacity retention >100 cycles.

References

[1] A. Kwade et al., Nature Energy 3(4) (2018) 290
[2] M. N. Obrovac et al., Chemical reviews, 114(23) (2014) 11444
[3] U. Kasavajjula et al., Journal of Power Sources 163 (2007) 1003
[4] Y. Jin et al., Journal of the American Chemical Society, 140(31) (2018) 9854
[5] N. Liu et al., Nature Nanotechnology, 9(3) (2014) 187
[6] F. Luo et al., Journal of The Electrochemical Society 162(14) (2015) A2509
[7] P.G. Karagiannidis et al., ACS nano, 11(3) (2017) 2742
[8] E. Greco et al., Journal of Materials Chemistry A 5(36) (2017) 19306