Dually cross-linked polyacrylic acid/graphene composite polymer as a toughened binder towards silicon electrode in lithium ion battery

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Silicon (Si) remains one of the most promising anode materials for next generation lithium-ion batteries, due to its abundance and high capacity (3579 mAh/g: almost 10 times higher than the capacity of graphite). The problems still facing Si-based battery commercialization are volume expansion, which results in rapid capacity fade, and continuous Li loss through SEI formation from electrolyte decomposition. One approach to address this issue is to use robust binders to maintain good contact between the active material and current collector by forming stable network within the electrodes.

This work will introduce a toughened binder system: a composite of polyacrylic acid and few-layered graphene (PAA-FLG). Polyacrylic acid (PAA) is an aqueous-based polymer, and possess a high concentration of carboxyl group, which can be effectively connected with other monomers via copolymerization, contributing to good adhesion. Graphene is well known for its good flexibility and electrical conductivity, which can improve the mechanical properties and the electron transfer within the electrode. A dually cross-linked PAA-FLG composite was formed by using Fe^{2+} ions and FLG as both cross linkers. It has been approved that Fe^{2+} ions could facilitate to create ionic cross-linking among PAA chains, and also could be cross-linked with the oxide layer on graphene through the oxygen functional groups[1]. Therefore, it is hypothesised that the dual cross-linked polymer PAA-FLG that coordinate through Fe^{2+} could provide enhanced tensile properties for the electrode to accommodate the large volume expansion of Si, and thus improve the longevity of cyclability.

Tensile test result shows that PAA-FLG indicates much better mechanical property in terms of stiffness and flexibility comparing with PAA polymers. A preliminary study was conducted on Si vs. Li/Li+ half cells with different binder systems: partially neutralized PAA and PAA-FLG, where PAA is used as the control binder. Cells are tested under the C rate of C/5. Electrochemical cycling data indicates that after first 100 charge-discharge cycles, the cells with PAA-FLG maintains high delithiation capacity (about 1380mAh/g), while the ones with PAA has dropped below 1100mAh/g. To further prove the hypothesis, a comprehensive and systematic study based on PAA-FLG will follow, including functional group investigation, adhesion and impedance testing.

References:
[1] M. Zhong, Y. Liu, and X. Xie, “Self-healable, super tough graphene oxide–poly(acrylic acid) nanocomposite hydrogels facilitated by dual cross-linking effects through dynamic ionic interactions,” J. Mater. Chem. B, vol. 3, pp. 4001–4008, 2015.