Plasma-Derived Si-Doped Graphene as an Anchoring Matrix for Silicon Nanoparticles: Enhancing Interfacial Stability in Li-Ion Anodes

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Silicon nanoparticles (Si NPs) have attracted significant attention as anode materials for Li-ion batteries (LiBs) due to their exceptional theoretical capacity (3579 mAh g^{-1}) [1]. However, their practical application is limited by severe volume changes and low electrical conductivity. Hybridizing Si NPs with graphene can mitigate these issues by buffering volume changes and improving conductivity, yet most graphene–Si NPs hybrids suffer from weak interfacial interactions, leading to loss of electrical contact, limited cycle life, and capacity fading.

Strengthening the graphene–Si NPs interface is therefore critical for improving anode lifespan. Heteroatom doping of graphene has been shown to bolster interfacial integrity by modifying binding energy and enhancing charge transfer, resulting in a more robust connection that suppresses agglomeration and improves cycling stability and efficiency [2,3]. Although silicon-doped graphene (SDG) has been investigated theoretically [4,5], experimental studies remain scarce, and no work has reported composites made of silicon-doped graphene and Si NPs (SDG-Si) or systematically examined the effect of Si doping of graphene on the structural integrity and electrochemical behaviour of graphene–Si anodes. Here, we aim to address this gap through a systematic experimental investigation of SDG–Si nanocomposites prepared by in-flight mixing in a double-reactor system. Silicon-doped graphene will be synthesized in a plasma reactor using ethanol as the graphene precursor and Tetramethylsilane (TMS) as silicon-doping source, while silicon nanoparticles will be generated separately by pyrolysis of monosilane (SiH_4) in a hot-wall reactor (Fig. 1).

We anticipate that Si-doped graphene will form a stronger interface with silicon nanoparticles than undoped few-layer graphene (FLG). Substitutional Si atoms protruding from the graphene lattice [6] can serve as superior anchoring sites; beyond merely increasing interfacial binding energy, they may provide a unique advantage by forming direct bridges with the nanoparticle surface to establish strong chemical bonding between Si NPs and SDG. This anchoring is expected to suppress interfacial SDG-Si NPs delamination under electrochemical and mechanical stress, enhancing both columbic efficiency and cycling life of the LIB anode. Moreover, doped graphene can exhibit higher specific capacity than pristine graphene [7], suggesting SDG–Si NPs composites may outperform FLG–Si systems at identical graphene loadings. Overall, this work provides fundamental insight into atomic-scale interface engineering for next-generation silicon anodes.

References

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Figures

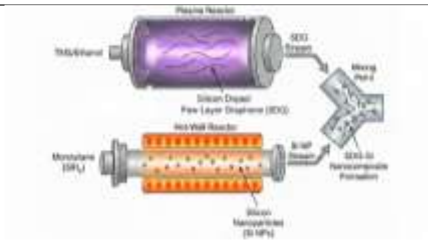

Figure 1: Schematic illustration of double reactor in-flight mixing system