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Superior lithium intercalation capacity of incommensurate graphene

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Graphite is the commonly used preferred anode in secondary batteries due to its high energy density, high power density, and high current efficiency, though with the limited theoretical capacity of 372 mAh/g. The limited capacity is caused by low diffusion of lithium within commensurately-stacked graphene layers due to the strength of repulsive interactions that arise due to the assembled nature of sp^2 carbon. The absence of commensurately-stacking order due results in a weaker Van der Waals forces so that the incommensurate multilayers can be considered as a single layer with modified electronic configuration^{1,2,3}. Here, we present a lithium intercalation study of novel graphene structure consisting up to 93% incommensurately-stacked layers. Raman Spectroscopy is used to study the stacking order and incommensurateness degree of graphene. Applying as an active material in lithium-ion battery cells, this incommensurate graphene demonstrates superior reversible capacity up to 1540 mAh/g. Structural and binding analyses studied by Raman, X-ray probing and Electron Energy Loss Spectroscopies reveal that lithium atoms highly intercalate within weakly-interacting graphene layers, followed by a further flexible rearrangement of layers for a long-term stable cycling. We propose a new model of Lithium insertion where capacity varies a number of graphene layers by $Li_{N+1}C_{2N}$ formula and it achieves 1674 mAh/g capacity in bi-layer configuration as a Li_3C_4 stoichiometry. In fact, the weakened interaction due to layers incommensurateness enables easy and full penetration of lithium, followed by flexible adjustment of the layers for stable long-term cycling. These findings promise the feasibility of the rapid development of light weight, cost-efficient secondary batteries with an increase of effective capacity by the factor of six compared to conventional graphite batteries.

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