Simple and novel electrografting monomer method to exfoliate HOPG for lithium-ion batteries

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Many methods have been developed in the last few decades to obtain high-quality graphene thin sheets. They are based on very different physicochemical processes. Here we demonstrate a novel one step and simple electrografting acrylate monomer method to exfoliate highly oriented pyrolytic graphite (HOPG) into thin nanoplatelets and even down to the single graphene sheet level. Among the high research activity in the area of surface modification, electrografting is a very powerful method which has received comparatively little attention. This is surprising because this technique has many attractive features for modification of conducting or semi-conducting surfaces. The main interest of the electrografting process is to solve the recurrent problem of the organic/substrates interface weakness. The electrografting warranties a robust polymer/substrates interface and offers the possibility to tailor the functionality of the grafted polymer film opening the door to a wide range of demanding technological applications.

The graphene sheets obtained through electrografting process give a stable suspension in dimethyl formamide (DMF), and they can self-precipitate on the surface of substrates after adding water as an antisolvent due to their strong surface hydrophobicity. Interestingly, the continuous films obtained exhibit ultratransparency (~98% transmittance), and the lateral size of the exfoliated graphene sheets observed by AFM ~1nm. Raman and TEM characterizations corroborate that the graphene sheets exfoliated by our electrochemical method preserve the intrinsic structure of grapheme and give preferentially monolayered graphene sheets.

The electrochemical behaviour of the acrylate monomer grafted graphene sheets was evaluated in lithium-half cells with no addition of conductive additive or binder. The PAN grafted graphene dispersed in DMF was coated on Cu foil and dried in a vacuum oven at 55°C for 12h. After 200 cycles, the reversible capacity was still kept at 300mAh/g at the current density of 50mA/g. These results indicate that the prepared high quality graphene sheets possess good electrochemical performances for lithium storage.

This work provides an efficient approach to obtain high-quality, cost-effective, and scalable production of "graphene ink", which may pave a way toward future applications in lithium-ion batteries.



Photo of the dispersed graphene sheets in a DMF solution, after electrochemical exfoliation of HOPG and a typical AFM image for graphene thin sheet cast on a MICA substrate.