Electrochemical hydrogen storage of graphene-based materials

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Quite recently, graphene, a conceptually new class of two-dimensional atomic crystal material, has attracted increasing interest for its unique structure, various peculiar properties, and a wide range of promising potential applications [1-2]. Efficient storage of hydrogen is not only an important issue of energy and environmental urgencies, but also a great challenge for material science research. C. Ataca et al. [3] theoretically predicted that Li atom covered graphene can serve as a high-capacity hydrogen storage medium with each decorated Li atom absorbing up to four H_2 molecules, amounting to a gravimetric density of 12.8 wt.%, hypothetically supposing that both sides of each graphene sheets are available. However, a small hydrogen adsorption(< 0.4 wt.%) was experimentally obtained for the pristine single-layer graphene sheets at cryogenic and room temperatures, and the authors attributed this small hydrogen adsorption to the serious agglomerates of the graphene sheets and a weak binding to hydrogen for pristine graphene sheets [4]. Therefore, besides keeping the grapheme separated, it is required to create more effective hydrogen absorption sites for graphene in order to further improve their hydrogen storage property.

Here we investigated the electrochemical hydrogen storage of graphene-based materials synthesized by two different methods－thermal expansion/exfoliation of graphite oxide and sonication exfoliation of graphite oxide followed by hydrazine reduction (Fig. 1). The thermal expansion derived graphene sheets show a higher hydrogen storage capacity of 145 mAh g^{-1} than that of sonication derived graphene sheets (125 mAh g^{-1}) due to the higher specific surface area. Further thermal reduction of the graphene samples leads to a decrease in the storage capacity, indicating that the presence of oxygen functional groups is benefit for the hydrogen absorption (Fig. 2). Moreover, we found that the electrocatalytic activity and hydrogen adsorption of the electrode of graphene sheets can be improved by decorating nickel nanoparticles on their surface. The graphene/nickel nanoparticle composite shows a capacity of 300 mAh g^{-1} (Fig. 3). However, a poor charge/discharge cycle performance is also observed, primarily due to the destruction of graphene-nickel interface structure and the anodic oxidation of the metallic nickel nanoparticles during the cycling process. We proposed a strategy to weaken the anodic oxidation of nickel particles and consequently improve the cycle performance of the composite electrode by tuning the discharge cutoff potential.

References:
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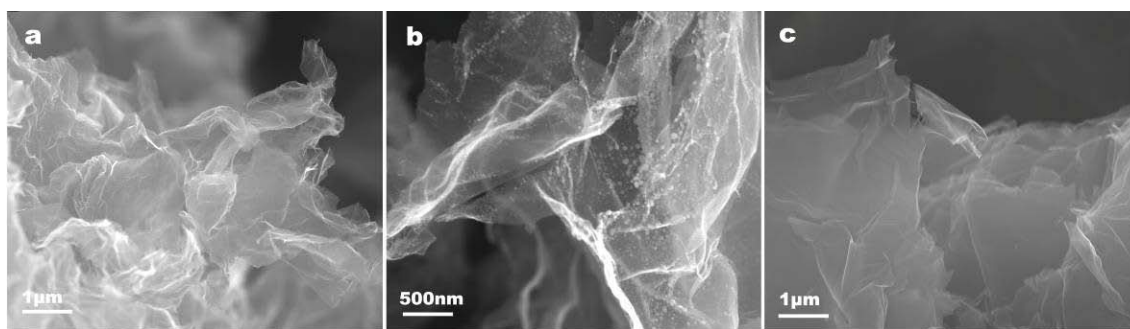
Fig. 1 – SEM images of (a) as-produced thermal expanded graphene sheets (TEGS); (b) graphene sheets decorated with nickel nanoparticles; (c) as-produced chemical reduced graphene sheets (CRGS).
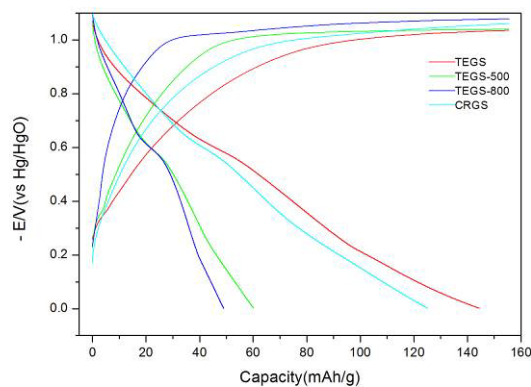

Fig. 2 – The initial charge-discharge curve of the electrode based on (a) as-produced thermal expanded graphene sheets (TEGS); TEGS powder reduced in hydrogen atmosphere at 500 °C (b) and 800 °C (c); (d) as-produced chemical reduced graphene sheets (CRGS).
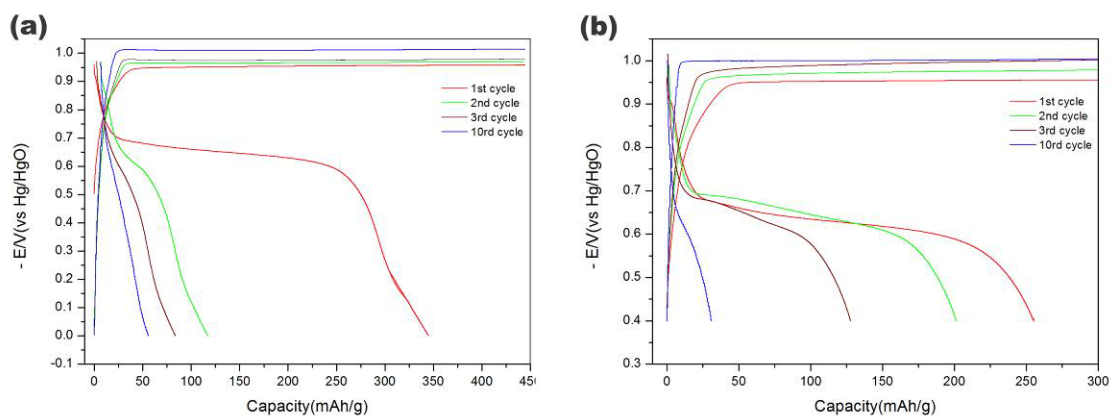

Fig. 3 – Charge–discharge curves of graphene-Ni electrodes at different cutoff potential: (a) 0 V vs. Hg/HgO and (b) -0.4 V vs. Hg/HgO.