Smart device, Energy storage

Synthesis of high-crystallinity randomly stacked graphene macrostructures

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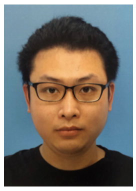


Abstract

In this study, we developed a method to synthesize highly crystalline and randomly stacked graphene macrostructures. By ultra-high temperature treatment at 1500–1800 C using ethanol as a reactive gas, graphene macrostructures with high crystallinity and high random stacking fractions were successfully synthesized from sponge-shaped graphene oxide (GO) assemblies formed by a freeze-drying method. Additionally, the random stacking fractions were further improved by adding cellulose nanofibers (CNF).

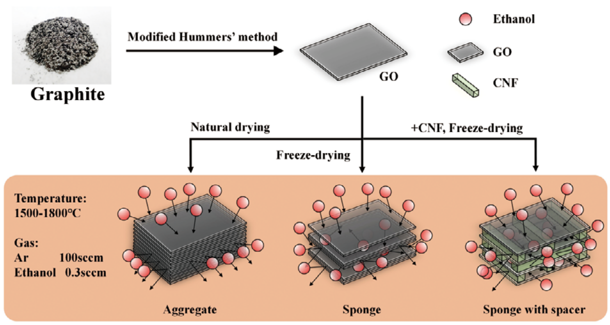
Background & Results

Graphene is a nanocarbon material with a variety of excellent properties, but its thinness and small size limit its application fields. Although graphene macrostructures are needed for macro-scale applications, a simple approach results in a structure similar to graphite and lost of the excellent properties of graphene. Assemblies with a high random stacking fractions prevent the degradation of properties caused by strong graphene interlayer interactions and enable the use of the excellent properties on the macroscale. The graphene sponge structures obtained in this study are expected to be applied in various devices including wearable strain sensors and battery electrodes.

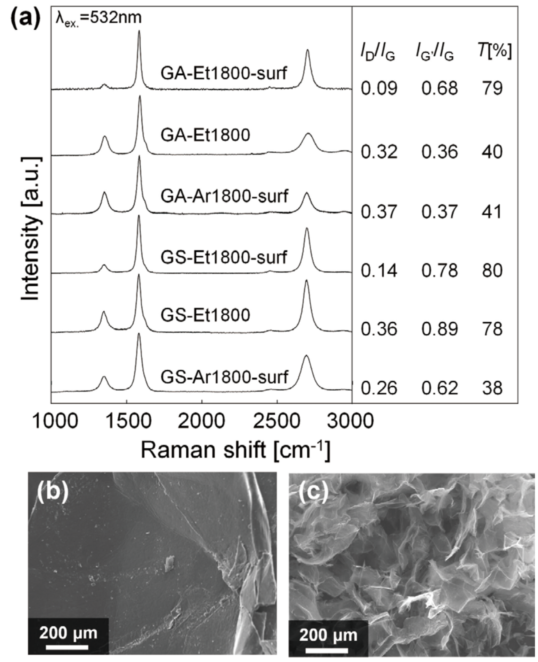
Significance of the research and Future perspective

Graphene is a nanocarbon material consisting of six-membered rings of carbon atoms with a thickness of one atom. Due to its unique electronic structure, graphene has excellent properties and is expected to find a variety of applications. However, since graphene is extremely thin and small, it is necessary to synthesize graphene macrostructures for macro-scale applications. For multilayer graphene, thermodynamically stable AB stacking structure is preferentially formed. Then, electronic structure of multilayer graphene is altered from that of single-layer graphene and approaches graphite due to strong interlayer interactions. On the other hand, the interlayer interaction is weak in a randomly stacked structure. To preserve the properties of graphene and enable its application on a macro scale, graphene macrostructures with high crystallinity and high random stacking fractions need to be synthesized.

In this study, we synthesized graphene macrostructure samples by various processes from graphene oxide (GO) and evaluated their crystallinity and random stacking fractions by Raman spectroscopy and other techniques. We found that the crystallinity and random stacking fractions of graphene microstructures were improved significantly when treated at ultra-high temperatures with ethanol as a reactive gas compared to when treated with inert argon. Furthermore, compared to the dense aggregated structures of GO as a starting material, sponge-like structures formed by freeze-drying resulted in higher crystallinity and higher random stacking fractions not only on the surface but also inside the macrostructures after the ultra-high temperature treatment. We consider that the ethanol-derived effects of carbon atom supply and defect etching can reach the interior of the GO sponges, resulting in efficient structural repair while preventing rearrangement of orientations between graphene layers.



Schematics of synthesis process of the graphene macrostructures.



(a) Raman spectra and (b,c) scanning electron microscope images of the graphene macrostructures.

Patent

Treatise Xu, Zizhao; Inoue, Taiki; Nishina, Yuta et al. Stacking Order Reduction in Multilayer Graphene by Inserting Nanospacers. Journal of Applied Physics. 2022, 132, p. 174305, doi: 10.1063/5.0103826
Xu, Zizhao; Nakamura, Shingo; Inoue, Taiki et al. Bulk-scale synthesis of randomly stacked graphene with high crystallinity, Carbon. 2021, 185, p. 368-375, doi: 10.1016/j.carbon.2021.09.034

URL http://www.ap.eng.osaka-u.ac.jp/nanomaterial/index.html

Keyword nanocarbon materials, reduced graphene oxide, interlayer interaction