



Facile, environmentally friendly, cost effective and scalable production of few-layered graphene



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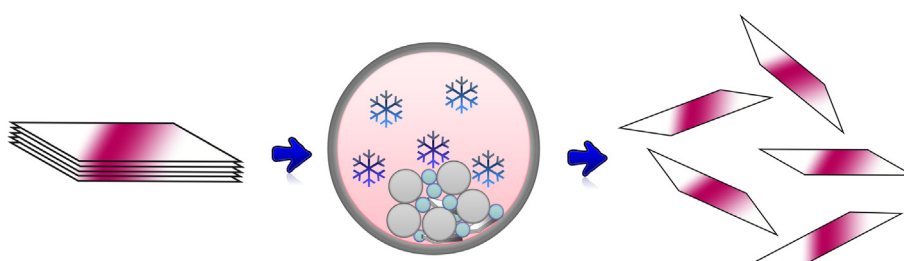
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HIGHLIGHTS

- A new method for large-scale production of mono- and few-layered graphene.
- Supercritical liquid phase shear-assisted exfoliation at extremely low temperatures.
- More than 61% of the flakes comprise less than 5 layers.
- Manipulating the surface energy of graphite to match it with that for several low melting point organic solvents.
- Presenting one of the fastest, cost effective and environmentally friendly exfoliation methods.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 24 March 2017

Received in revised form 19 May 2017

Accepted 9 June 2017

Available online 10 June 2017

Keywords:

Liquid phase exfoliation

Graphene

Shear-assisted exfoliation

Scalable method

Nitrogen

ABSTRACT

Commercialization of graphene is still one of the biggest challenges in the carbon field despite the development of several methods for its production. The lack of simple, cost-effective and scalable methods for mass-production of graphene hampers its promotion to the market. Here, we propose a new method for large-scale production of mono- and few-layered graphene via liquid phase exfoliation with the use of wet ball milling in the presence of organic solvents at extremely low temperatures. The wet ball milling combined with the temperature modulated high surface energy solvents affords exfoliation of bulk graphite into graphenes in a fast, scalable, cost effective and environmentally friendly process. The thorough statistical analysis of as-prepared graphene flakes demonstrates that more than 61% of the flakes comprise less than 5 layers, while ~14% of the flakes were monolayer graphene. Combined with the ~30% yield of few-layer graphene out of the graphite precursor, this method demonstrates incredible efficiency in just 45 min. In the presence of methanol, our method results in formation of predominantly bi-layer graphene, which is more difficult to obtain in scalable fashion, than mono-layer graphene. The high quality of as-obtained graphenes is fully confirmed by Raman spectroscopy, TEM, SAED, AFM and X-ray photoelectron spectroscopy.

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Abbreviations: TEM, transmission electron microscopy; XPS, X-ray photoelectron spectroscopy; SEM, scanning electron microscopy; SAED, select area electron diffraction; AFM, atomic-force microscopy; GNPs, graphene nanoplatelets; CNT, carbon nanotubes; MGNP, methanol-exfoliated GNP; EGNP, ethanol-exfoliated GNP; AcGNP, acetone-exfoliated GNP; DEEGNP, diethyl ether-exfoliated GNP; ISPGNP, isopropyl alcohol-exfoliated GNP; DMF, *N,N*-dimethylformamide; NGNP, non-organic solvent-exfoliated GNP; NMP, *N*-methyl-2-pyrrolidone.

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<http://dx.doi.org/10.1016/j.cej.2017.06.046>

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