

Supplementary Information

Single Layer Nano Graphene Platelets Derived From Graphite Nanofibres

*Kai Huang,^{a,b} Géraud Delport,^c Lucile Oricin-Chaix,^c Carlos Drummond,^{a,b} Jean-Sebastien Lauret,^c and Alain Penicaud^{*a,b}*

^a CNRS, Centre de Recherche Paul Pascal (CRPP), UPR 8641, F-33600 Pessac, France.

^b Université Bordeaux, CRPP, UPR 8641, F-33600 Pessac, France

^c Laboratoire Aimé Cotton, CNRS, ENS Cachan, Université Paris Sud, campus d'Orsay, bât 505 91405 Orsay cedex France.

E-mail: penicaud@crpp-bordeaux.cnrs.fr

Contents :

Fig. S1 Raman spectrum of starting GNF nanofibre material.

Fig. S2 SEM and TEM images of starting GNF nanofibre material.

Fig. S3 Additional TEM images of starting GNF nanofibre material.

Fig. S4 XPS spectrum of starting GNF nanofibre material.

Fig. S5 XPS spectrum of C 1s core level signal of starting GNF nanofibre material.

Fig. S6 XPS spectra of C 1s core level signal with fitting peaks of GNFIC/THF fresh and 6-month-aged solutions.

Table S1. Deconvolution result of XPS C 1s signal of GNFIC/THF fresh and 6-month-aged solutions.

Fig. S7 Photoluminescence spectra of the GIC/NMP (i.e. from graphite rather than GNFs) fresh solution at t~405min after exposure to air.

Additional experiments on alternative GNF sources:

Fig. S8 The ID/IG ratio distribution of GNFIC (from source of GNF1).

Fig. S9 The ID/IG ratio distribution of GNFIC/THF (from source of GNF3).

Fig. S10 AFM images of GNFIC/NMP solution (source of GNF1) with height and lateral size distributions.

Fig. S11 AFM images of GNFIC/NMP solution (source of GNF4) with height and lateral size distributions.

Recommended manipulation for all processes using potassium and dealing with potassium waste.

Alkali metals are strong reducing agents that react violently with water. Inexperienced users should not attempt to perform reduction reactions with potassium metal. Laboratory, that work with alkali metals should be equipped with fire extinguisher for metals. Do not use CO₂ fire extinguisher. Pay attention to the possible formation of KO₂, that is formed on the surface of the potassium metal due to the reaction with oxygen upon prolonged storage. This material may explode when it is exposed to pressure - e.g. cutting.

Procedure : A chunk of potassium metal is cut to an approximate size of 5 x 5 x 10 mm³ into its storage oil and then placed into moisture free hexane under a clean fume hood. It is then placed in a vial and taken into the glove box, that is used as stock material. Manipulating potassium and potassium intercalation compounds in an inert atmosphere glove box is less dangerous but users should pay attention to potassium waste. Cleaning of the GNFIC solutions coated surfaces can be handled in fume hood and is less of a danger due to the tiny amount of reducing agent contained in the solutions. Potassium waste handling should be done in a clean fume hood, with care. Potassium chunks (of size < 1 mm³) should be reacted progressively with butanol, isopropanol, ethanol, methanol, and water. Waste paper and tools stained with potassium should be immersed carefully into ethanol. If larger quantities of potassium have to be dealt with, a more systematic set-up has to be used with a cooling system.¹

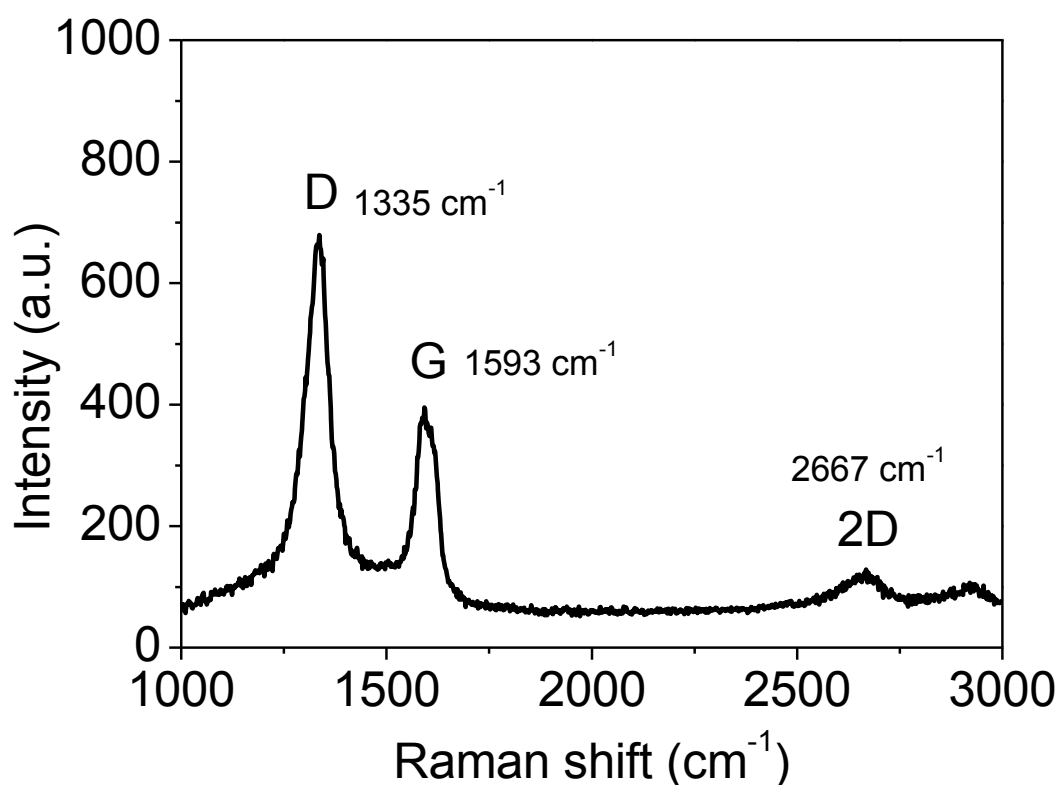
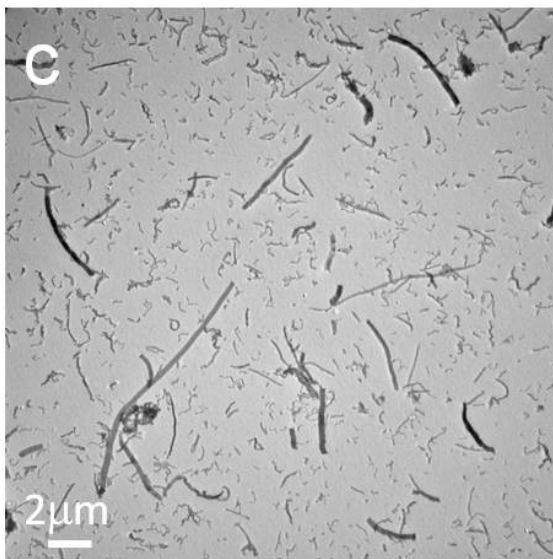
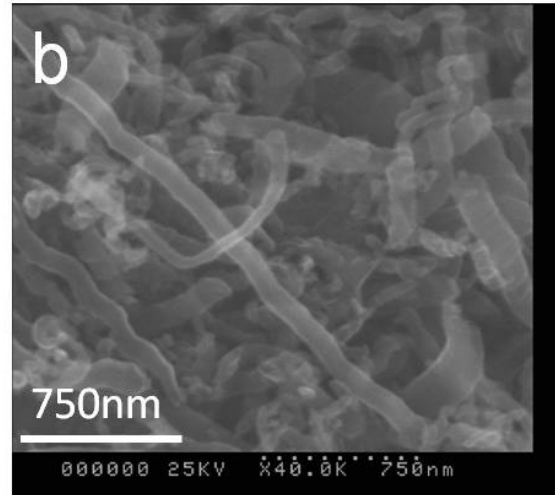
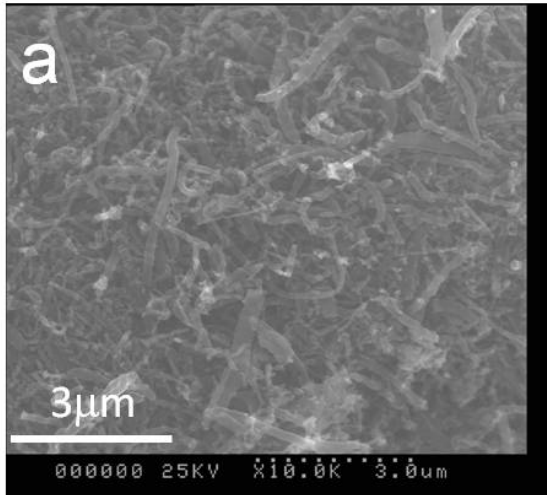
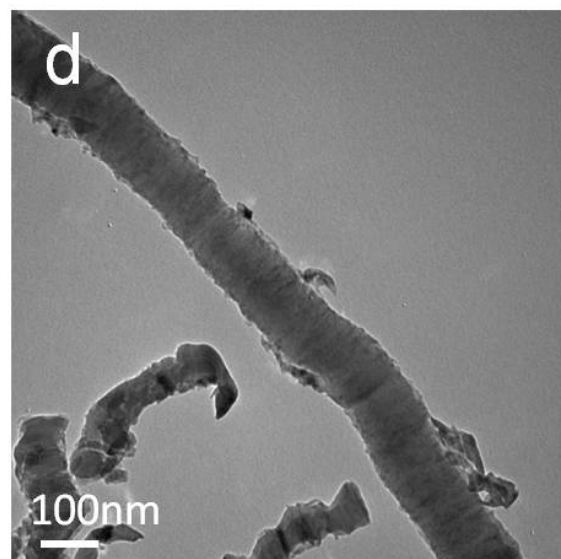


Fig. S1 Raman spectrum of starting nanofibre material using $\lambda = 638\text{nm}$ laser.



2 microns
HV=75,0kV
Direct Mag: 4000x
AMT Camera System



100 nm
HV=75,0kV
Direct Mag: 100000x
AMT Camera System

Fig. S2 (a, b) SEM and (c, d) TEM images of starting nanofibre material.

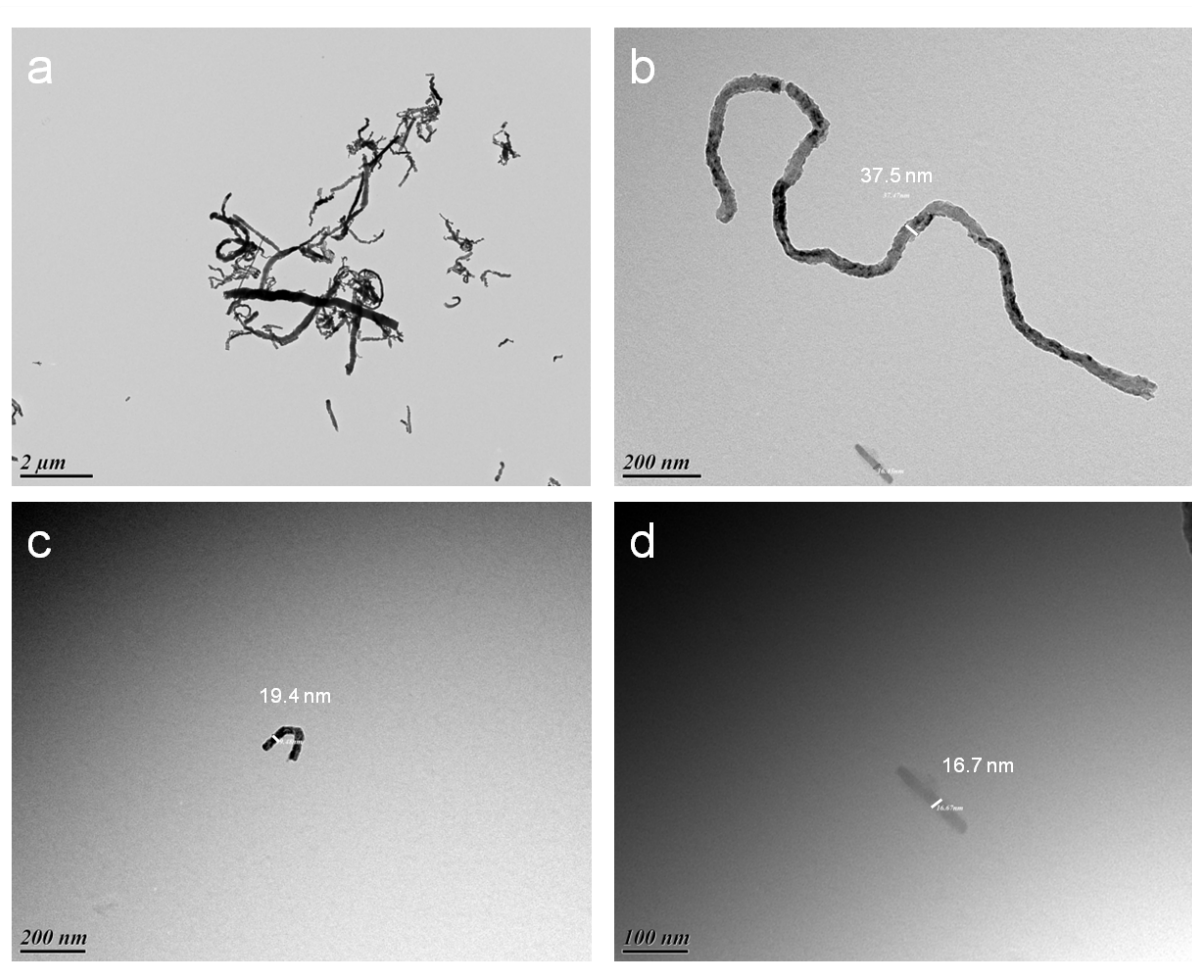


Fig. S3 (a-d) additional TEM images of starting nanofibre material. Although there is a wide diameter distribution, small lateral size GNF are indicated can be seen in (a) and are indicated in (b) to (d).

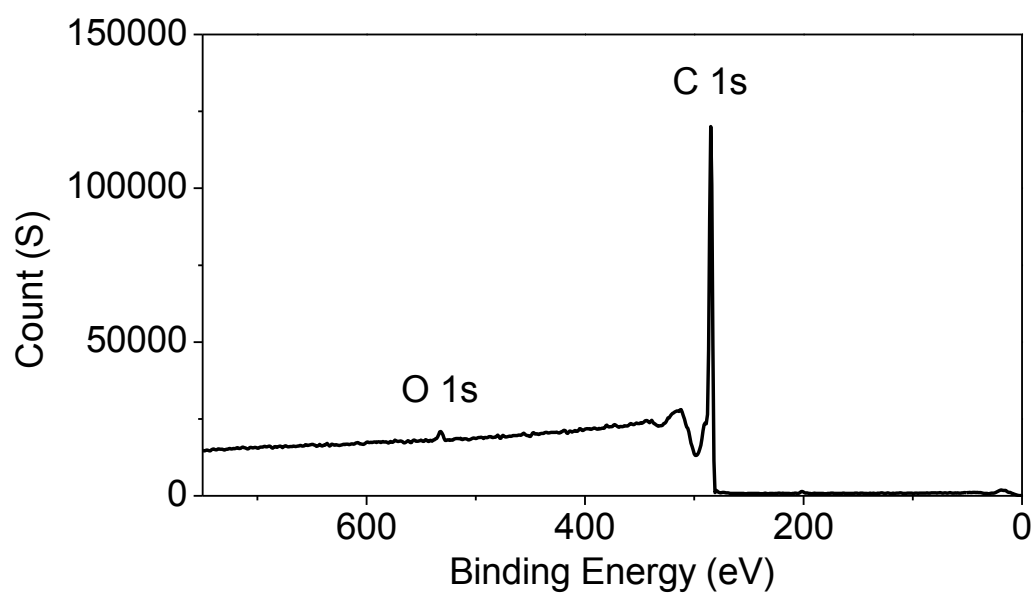


Fig. S4 XPS spectra of starting nanofibre material.

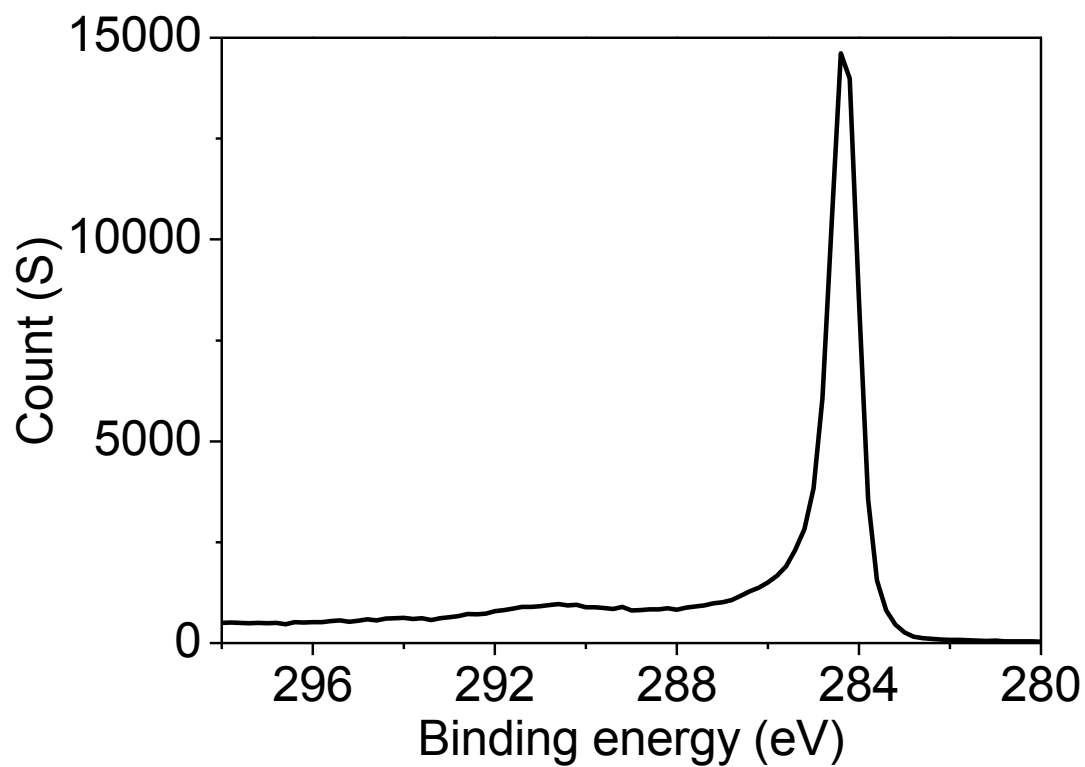


Fig. S5 XPS spectrum of C 1s core level signal of starting nanofibre material.

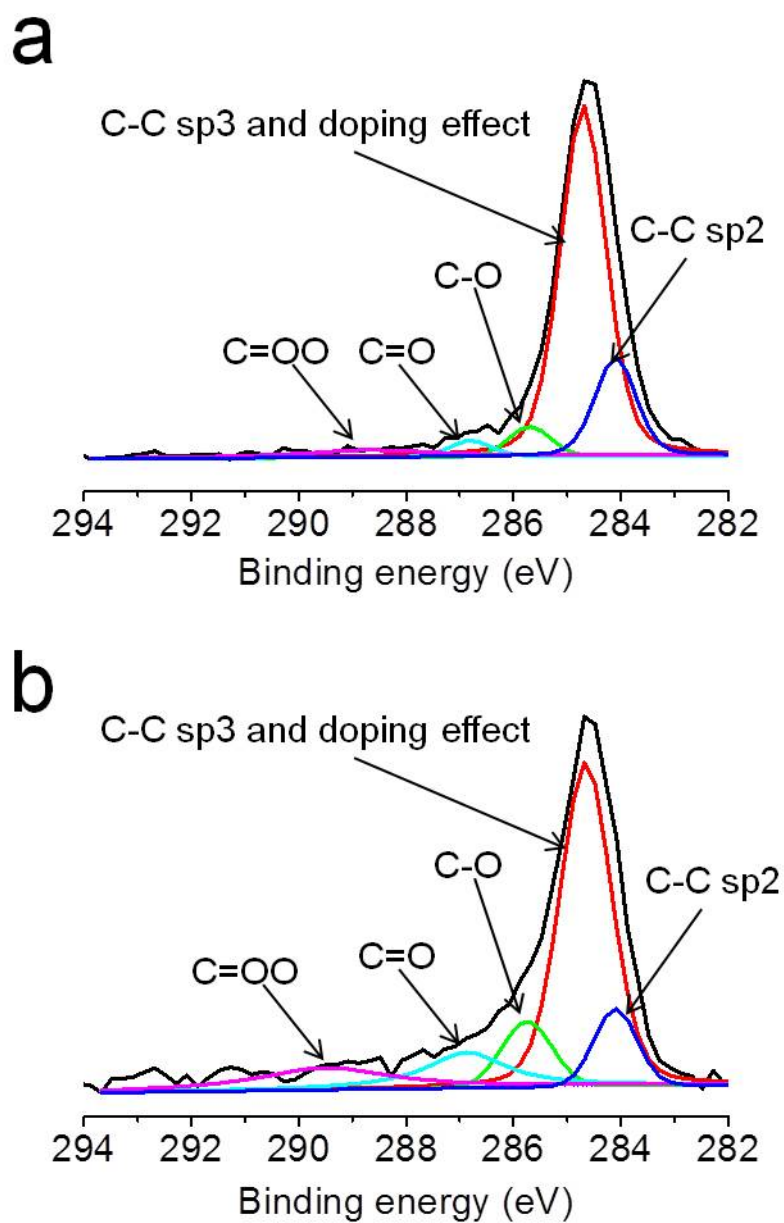


Fig. S6 XPS spectra of C 1s core level signal of (a) GNFIC/THF fresh solution and (b) GNFIC/THF 6-month-aged solution.

Table S1. Deconvolution result of XPS C 1s signal of GNFIC/THF fresh solution and 6-month-aged solution.

	GNFIC/THF fresh solution		GNFIC/THF 6-month-aged solution	
	Position (eV)	Percentage (%)	Position (eV)	Percentage (%)
C-C sp ²	284.1	17.3	284.1	9.7
C-C sp ³	284.7	69.7	284.7	54.7
C-O	285.7	4.7	285.8	9.8
C=O	286.8	2.7	286.9	12.7
C=OO	288.7	5.6	289.5	13.1

The C1s XPS signals of GNFIC/THF fresh solution and 6-month-aged solution can be fitted by five components but mainly dominated by C-C sp² peak at 284.1 eV and C-C sp³ peak at 284.7 eV. These two peaks are in good agreement with the HOPG sp² carbon and diamond-like sp³ carbon in the literature.²⁻⁴ The other peaks at higher binding energy are attributed to carbon atoms bonded with oxygen functionalities as C-O single bond, C=O double bond and C=OO bond.⁵

The deconvolution results are also shown in Table S1 with peak position and percentage. The ratio of sp³/sp² increased to 5.64 for the 6-month-aged solution compared to 4.03 for the fresh solution. The total oxygen bonded carbon increased from 13 % in the fresh solution to 35.6% for the 6-month-aged solution. These results are in agreement with the Raman study showing ageing of the THF GNFIC solutions

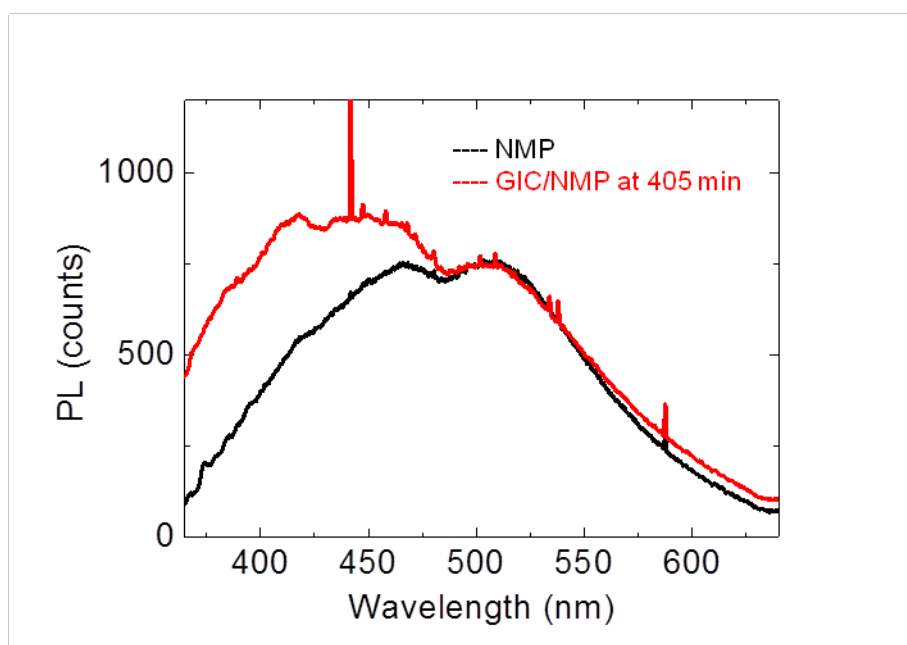


Fig. S7 Photoluminescence spectra of the GIC/NMP (i.e. from graphite rather than GNFs) fresh solution at t~405min after exposure to air.

Additional experiments on alternative GNF sources.

- GNF1 : GNF from CATALYX.
- GNF3 : GNF from STREM (acid washed). Catalog number : 06-0170
- GNF4 : GNF from STREM (heat treated). Catalog number : 06-0172.

AFM and Raman data are presented below. Exfoliation of the GNFs to calibrated graphene platelets is observed in all cases.

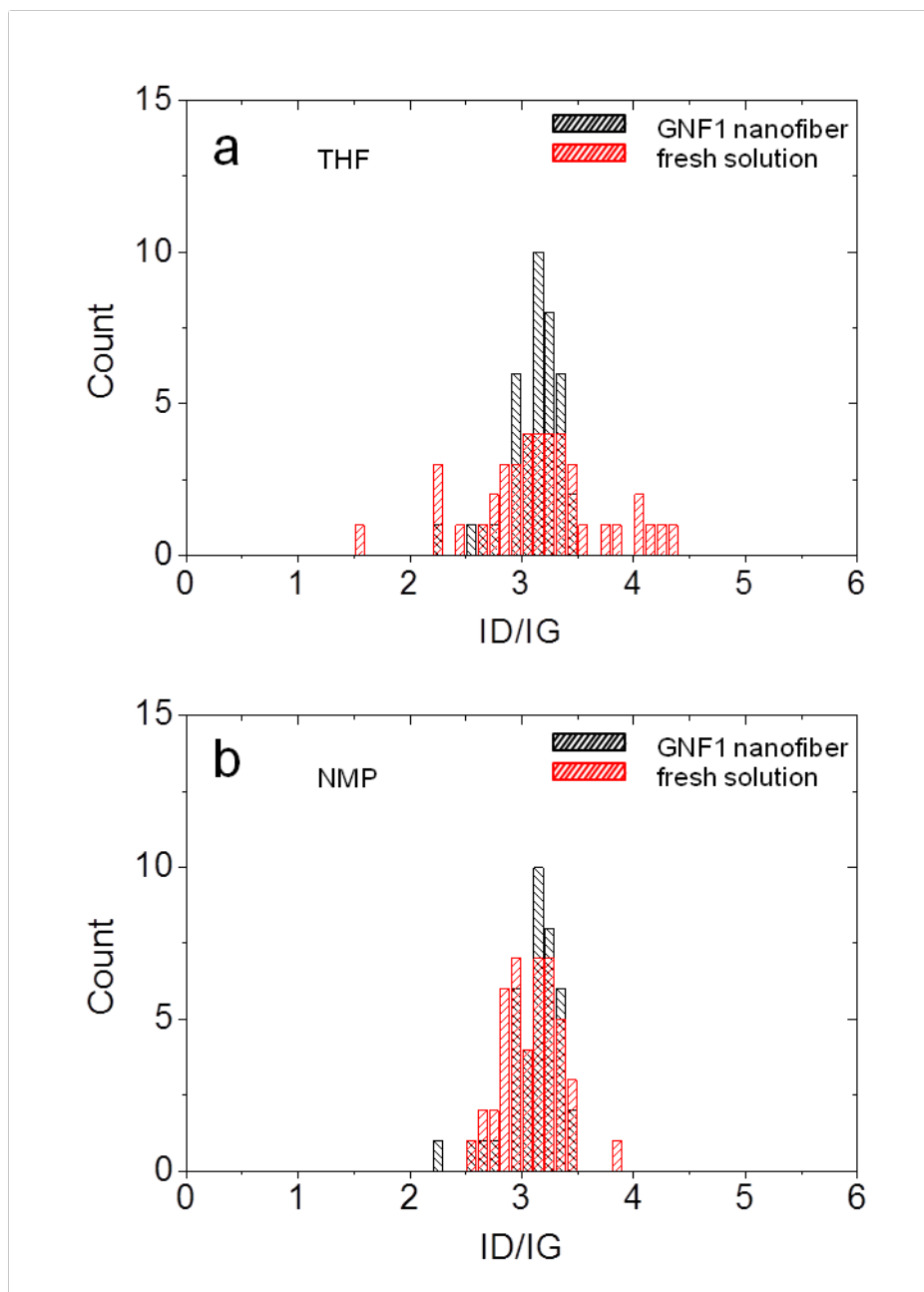


Fig. S8 The ID/IG ratio distribution of GNF1C (from source of GNF1) in THF (a) and NMP (b). The GNF1 starting nanofibre was also recorded and the result is shown in black.

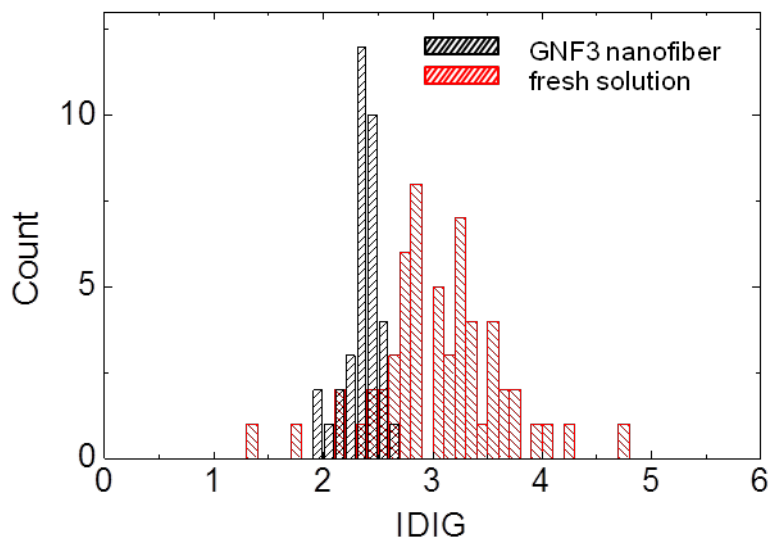


Fig. S9 The ID/IG ratio distribution of GNFIC/THF (from source of GNF3). The GNF3 starting nanofibre was also recorded and the result is shown in black.

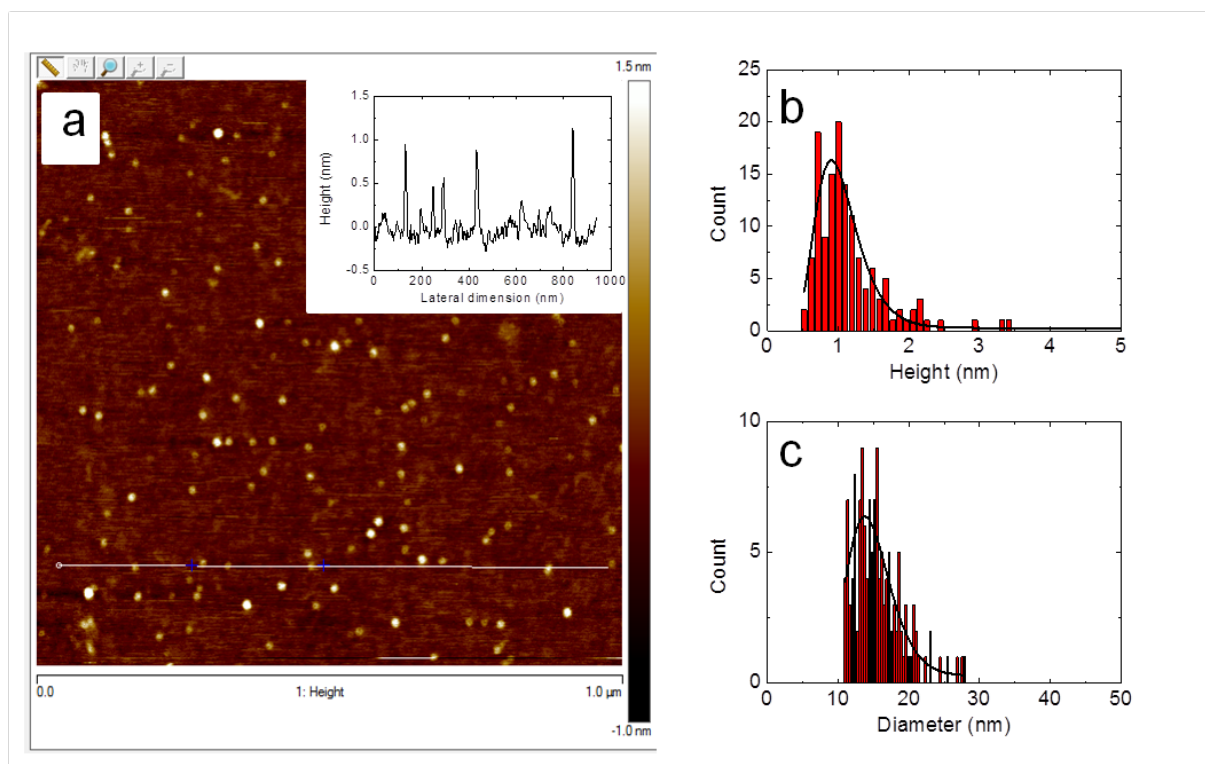


Fig. S10 (a) AFM topography images of deposits of GNFIC/NMP solution (source of GNF1) on freshly cleaved mica surfaces. Cross sections of the topography images taken along the white line are plotted in inserted figure, showing that the height of the nano objects on the surface is less than 1 nm. (b) Height distribution of nanographene; the mean height of the objects is 1 nm. (c) Lateral size distribution of nanographenes; the mean size is 14.5 nm before tip deconvolution. Lines in (b) and (c) correspond to a log normal distribution fit.

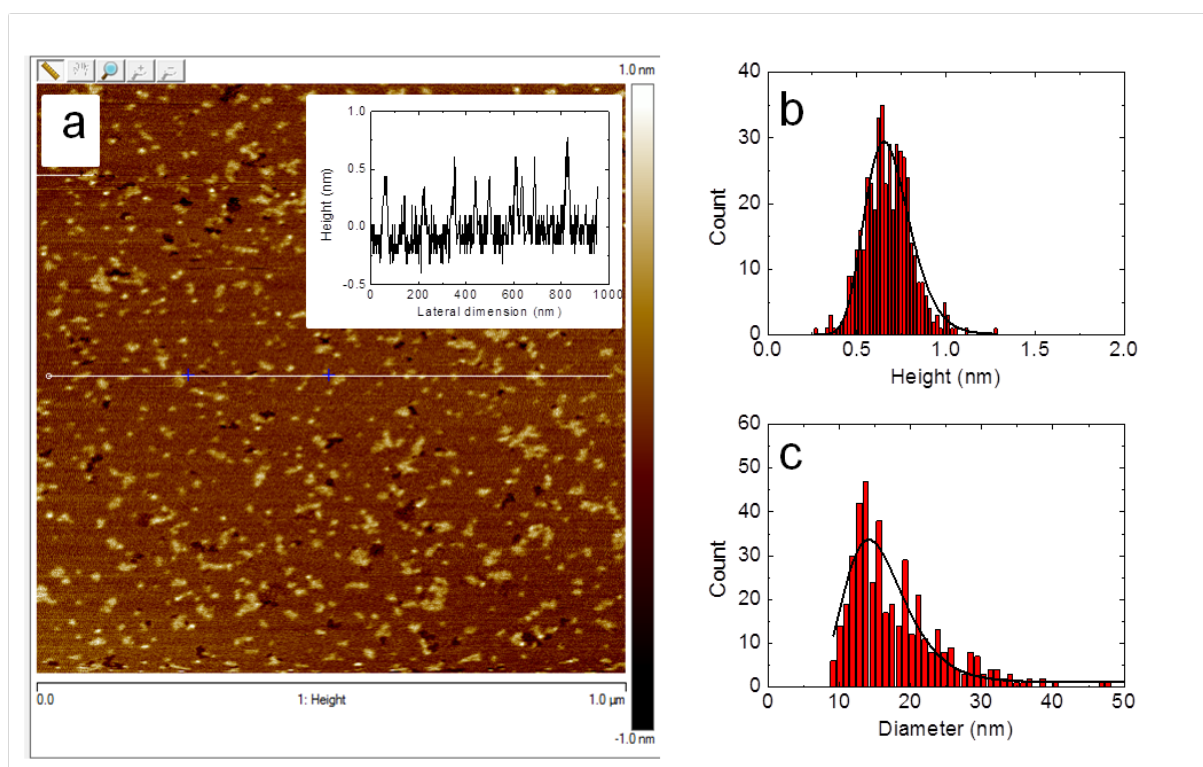


Fig. S11 (a) AFM topography images of deposits of GNF4/THF solution (source of GNF4) on freshly cleaved mica surfaces. Cross sections of the topography images taken along the white line are plotted in inserted figure, showing that the height of the nano objects on the surface is less than 1 nm. (b) Height distribution of nanographene; the mean height of the objects is 0.7 nm. (c) Lateral size distribution of nanographenes; the mean size is 15 nm before tip deconvolution. Lines in (b) and (c) correspond to a log normal distribution fit.

References

- 1) M.A. Armour, Hazardous Laboratory Chemicals Disposal Guide, Third Edition, Lewis Publishers 2003, p. 414.
- 2) J. F. Morar, F. J. Himpsel, G. Hollinger, J. L. Jordan, G. Hughes, F. R. McFeely, *Phys. Rev. B*, 1986, **33**, 1340.
- 3) J. A. Johnson, D. Holland, J. B. Woodford, A. Zinovev, I. A. Gee, O. L. Eryilmaz, A. Erdemir, *Diamond Relat. Mater.* 2007, **16**, 209.
- 4) M. J. Webb, P. Palmgren, P. Pal, O. Karis, H. Grennberg, *Carbon*, 2011, **49**, 3242.
- 5) G. Beamson, D. Briggs, High Resolution XPS of Organic Polymers - The Scienta ESCA300 Database, Wiley Interscience, 1992.