Supporting Information for:

Optoelectronic properties of graphene thin films deposited by Langmuir-Blodgett assembly

HoKwon Kim[a*], Cecilia Mattevi[a*], Hyun Jun Kim[b], Anudha Mittal[c], K. Andre Mkhoyan[c], Richard E. Riman[b], Manish Chhowalla[b]

[a]CASC, Department of Materials, Imperial College London, London, SW7 2AZ, United Kingdom
[b]Department of Materials Science and Engineering, Rutgers University, Piscataway, New Jersey 08854, United States of America
[c]Department of Chemical Engineering & Materials Science, University of Minnesota, 151 Amundson Hall, 421 Washington Avenue SE, Minnesota 55455, USA

*Emails: hknano@gmail.com, c.mattevi@imperial.ac.uk

Absorption Coefficient Calculations

Using the Fresnel's equation for thin film limit the optical transmission for mono-to-few-layered graphene films can be written as[1-3]:

$$T = \frac{1}{\left(1 + \frac{\sigma_{op}^{G} t}{2\varepsilon_0 c}\right)^2}$$

For pristine graphene, $\sigma_{op}^{G} = \frac{G_0}{a} = \frac{e^2}{4\hbar a} = \frac{6.08\times10^{-5}\,\Omega^{-1}}{3.35\times10^{-10}\,\mathrm{m}} = 181,450\ \Omega^{-1}\mathrm{m}^{-1}$ where $a \sim 3.35$ Å is the interlayer spacing between the graphene layers which was assumed as the thickness of a monolayer.

Then the absorption coefficient of pristine graphene is[4]:

$$\alpha \approx \frac{Z_0 \sigma_{op}}{2} = \frac{376.7\Omega \times 181,450\ \Omega^{-1}\mathrm{m}^{-1}}{2} = 0.0342\ \mathrm{nm}^{-1}$$

By fitting the above equation to the experimental transmission versus thickness plot (**Figure 5b**), the optical conductivity of Langmuir-Blodgett film from this work was calculated to be 40,241 $\Omega^{-1}\mathrm{m}^{-1}$.

The absorption coefficient is then,

$$\alpha \approx \frac{Z_0 \sigma_{op}}{2} = \frac{376.7\Omega \times 40,241\ \Omega^{-1}\mathrm{m}^{-1}}{2} = 0.0076\ \mathrm{nm}^{-1}$$

This is a much smaller value than the absorption coefficient of an ideal graphene film.

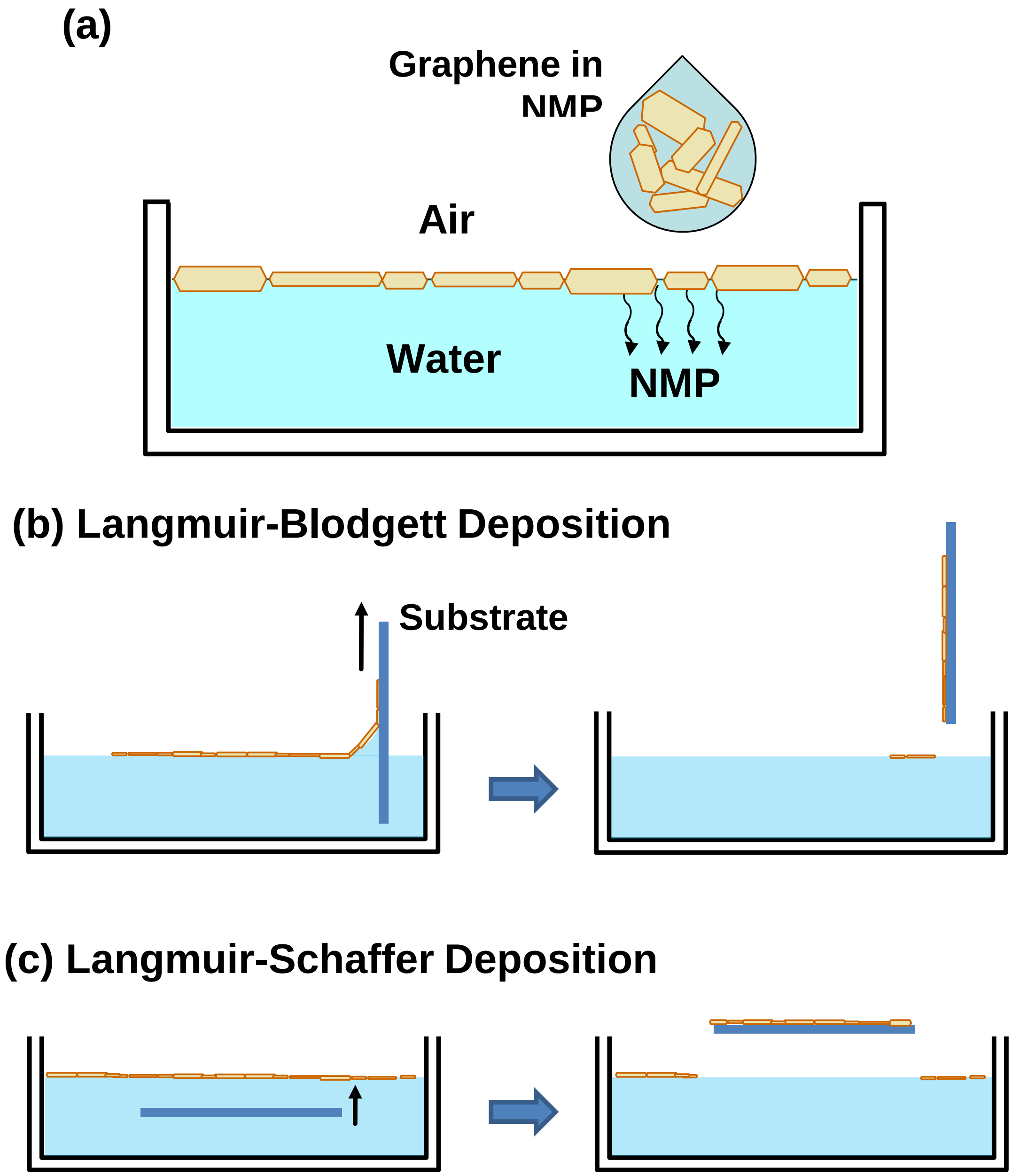

Figure S1 Schematic of film deposition processes using Langmuir-Blodgett assembly at the air-water interface. a) When dispersion of graphene/NMP is dropped onto a bath of water, Langmuir-Blodgett film forms on the surface of water as graphene is immiscible with water while NMP readily dissolves in water. After the formation of a Langmuir-Blodgett film, the film can be deposited on a pre-immersed substrate by Langmuir-Blodgett Deposition (b) or Langmuir-Schaffer Deposition (c).

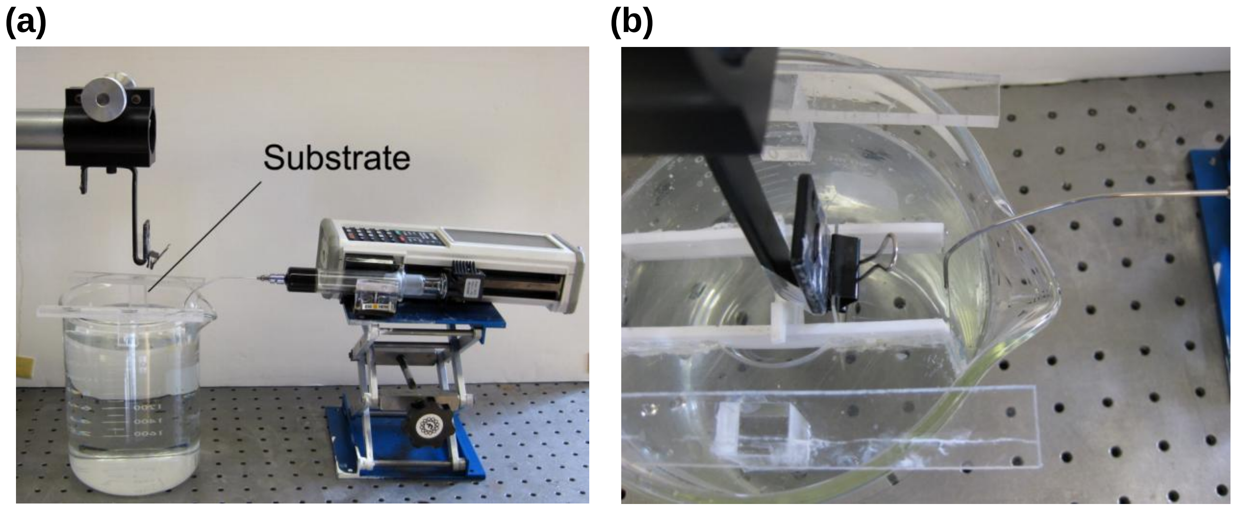

Figure S2 Photographs of the automated Langmuir-Blodgett deposition/dip-coater assembly. a) Side view of the dip-coater setup. b) Close-up, top view of the dip-coater setup. The substrate (glass slide) immersed in the custom-made trough is clamped to the automatically actuated arm that slowly pulls the substrate vertically up, while the injection of graphene/NMP dispersion onto the surface of water within the trough was controlled by the automated syringe pump. The trough is constructed by the walls made of Teflon and Plexiglas.

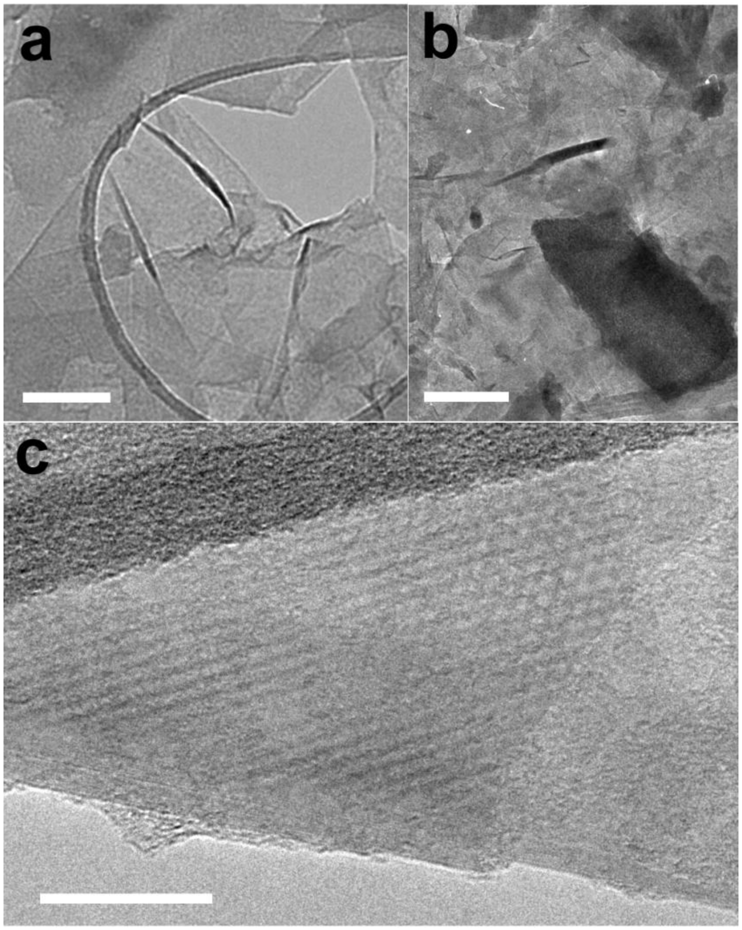

Figure S3 Bright field TEM images of exfoliated graphene flakes from GNMP1 and its centrifugation sediments. a) Exfoliated graphene flakes of supernatant suspension which was drop-cast on TEM grid. Scale bar: 100 nm. b) Sediment obtained after final centrifugation step for GNMP1. Scale bar: 200 nm. c) HRTEM image of a FLG. Scale bar: 2 nm.

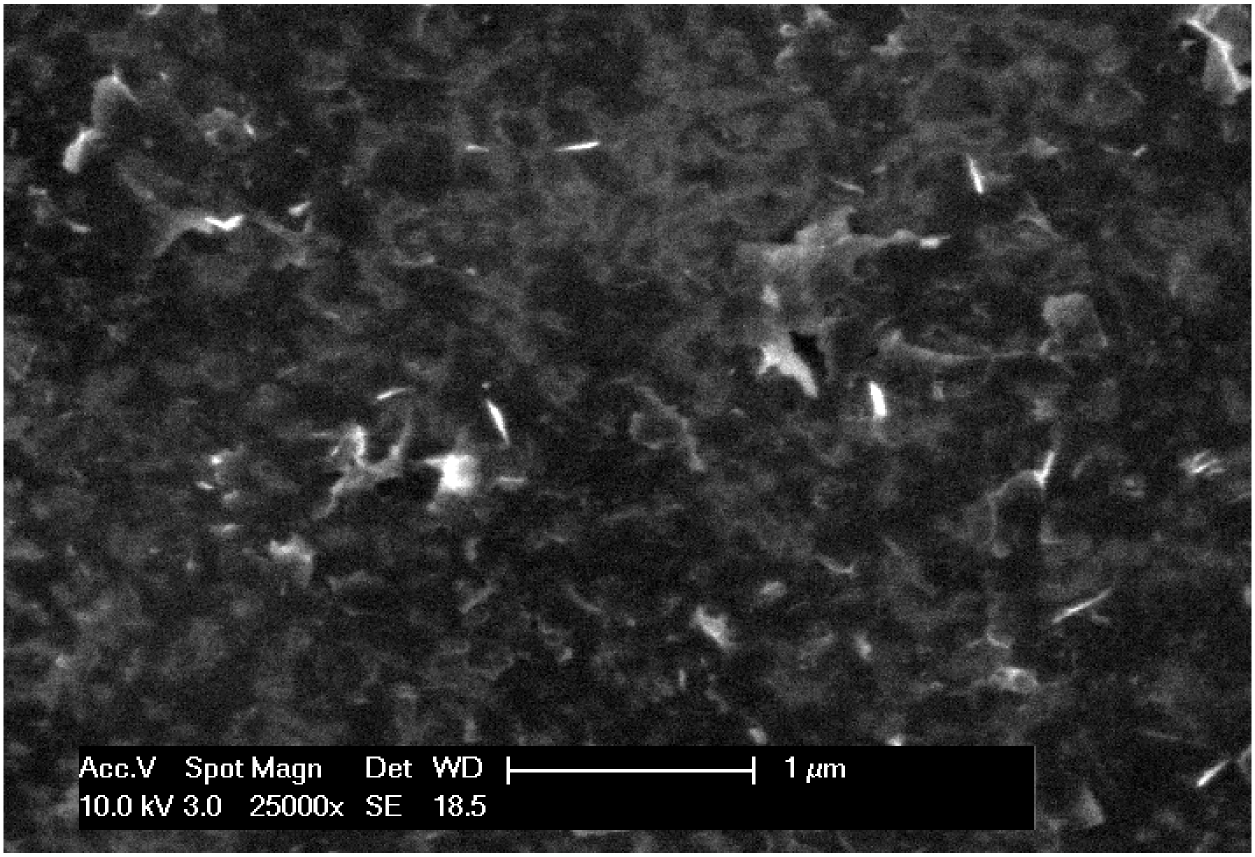

Figure S4 SEM image of doubly deposited LB film (from GNMP1).

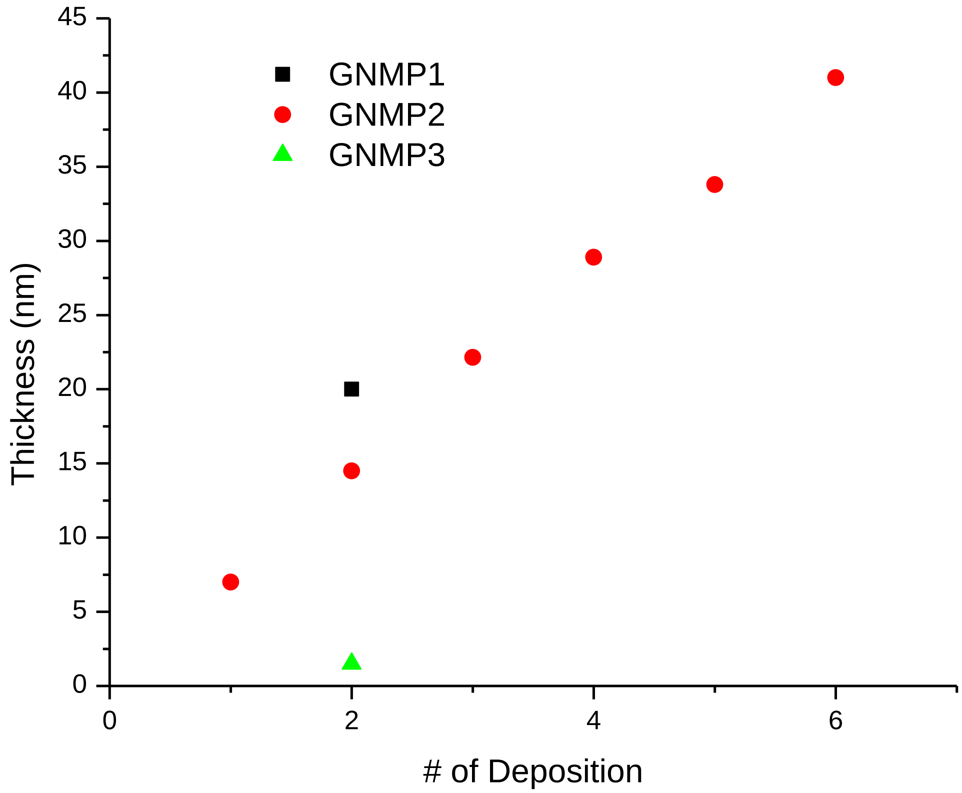

Figure S5 Thin film thickness versus number of LB deposition of self-assembled graphene films.

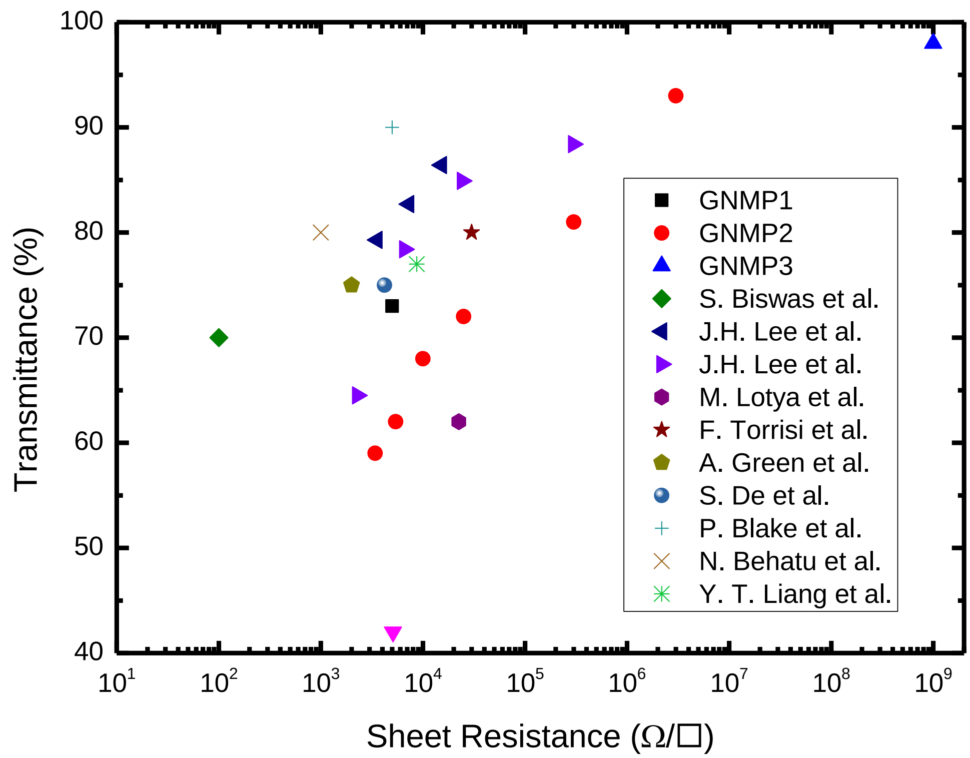

Figure S6 Transmittance vs. Sheet Resistance graph for LB Films for different dispersion types (GNMP1, GNMP2, GNMP3) and published values from literature for unfunctionalized graphene. (Y. Hernandez et al.[5] , Biswas et al.[6], Lee et al.[7], Lotya et al.[8], Torrisi et al.[9], Green et al.[10], De et al.[11], Blake et al.[12], Behatu et al.[13], and Y. T. Liang et al.[14])

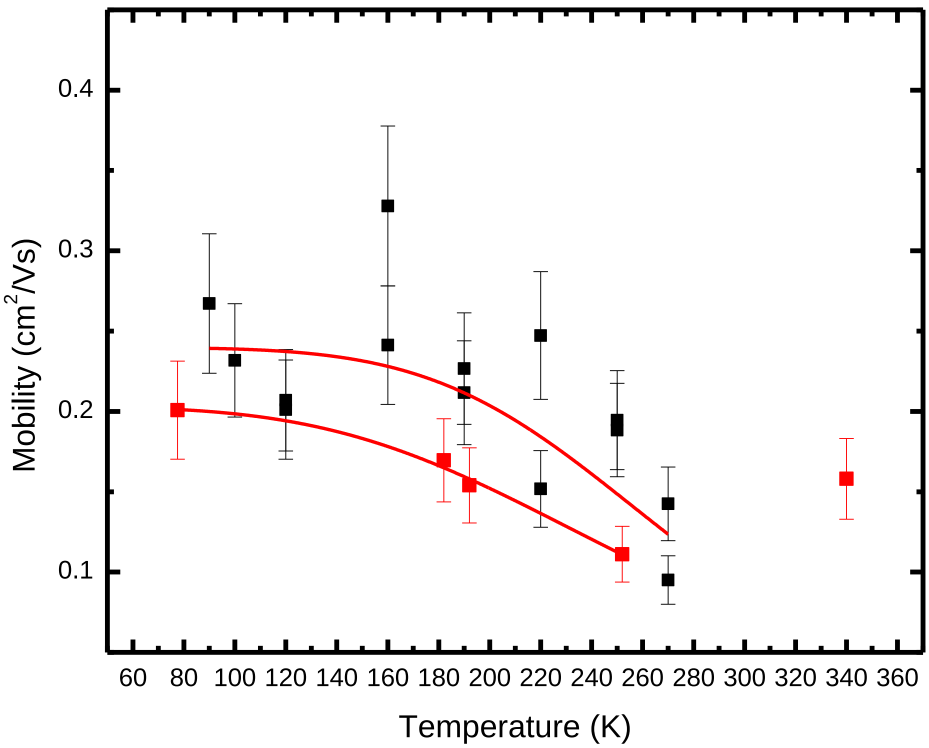

Figure S7 Temperature dependent hole mobility from two different devices fabricated from GNMP2. The solid curves are fitted based on Equation 5.

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