## **Supporting Information to:**

## **Critical Parameters in Exfoliating Graphite into Graphene**

## Matat Buzaglo, Michael Shtein, Sivan Kober, Robert Lovrincic, Ayelet Vilan and Oren Regev

- S1) a schematic description of the different surfactants employed for dispersing GS;
- ${\bf S2}{\bf )}$  Calibration plots of TS and BS power
- $\mathbf{S3}$ ) effect of dispersant to graphite ratio;
- 10 S4) UV-vis spectra of GS:TX-100 solution and TX-100 alone;
- S5) Thermograms of TX-100 and Graphite
- **S6**) Stability of GS dispersion;
- S7) Dispersant type versus graphene concentration and measured solvent surface tension
- S8)Detailed Pluronic composition and resulting GS concentration;
- $_{15}$  S9) GS average (characteristic) length
  - S10) Raman spectrum 2D band Lorentzian line fit analysis for GS characterization
  - S11) AFM images;

Finally, Table S1 compares the dispersing parameters and resulting GS concentrations between this work and some former ones.

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- (a) Cetyltrimethylammonium bromide (CTAB), cationic surfactant, n=7
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- (b) Didodecylammonium Bromide (DDAB), cationic surfactant, n=5
   (c) Sodium dodecyl sulfate
- (SDS), anionic 40surfactant, n=5
- (d) sodium cholate hydrate (SC)anionic surfactant
- (e) *Triton X-100* (*TX-100*) nonionic surfactant, n=9-10
- (f) Pluronic®s n & m change (See Fig. S8)



**Figure S2:** Calibration plots of (a) BS and (b) TS power. The temperature was measured in an isolated vessel filled with 10mL <sup>65</sup> DI water. The calculated power inputs were calculated based on the resulted slopes values (Power= Slope\*Weight\*Average heat Capacity)

Figure S1. Graphene dispersants used in this study.

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The supporting information includes the following figures:



**Figure S3.** Example of optimization sets (a) GS-CTAB and (b) GS-SDS dispersions, prepared by bath sonication for 3 hour. Each set contains 3 vials with GS: surfactant ratios of: 1:2, 1:1 & 5 2:1 – left to right.



**Figure S4.** UV-Vis absorption spectrum of GS-TX-100 dispersion, prepared by bath sonication (full line), and aqueous <sup>10</sup> solution of 5 mg/mL TX-100 (dashed line).



**Figure S5(a):** Thermogram of pure TX-100. The blue curve indicates the sample weight-loss in the ordinate (left Y-axis) with respect to time or temperature (abscissa) upon heating to 400°C. <sup>55</sup> The red curve is the time derivative of the weight-loss curve (mg/sec, right Y-axis). The dashed line is a stepwise approximation to the weight-loss is due to dispersant removal.



<sup>30</sup> Figure S5(b): Thermogram of pure Graphite. The blue curve indicates the sample weight-loss in the ordinate (left Y-axis) with respect to time or temperature (abscissa) upon heating to 400°C. The red curve is the time derivative of the weight-loss curve (mg/sec, right Y-axis). The dashed line is a stepwise
 <sup>35</sup> approximation to the weight-loss is due graphite oxidative decarbonylation.



**Figure S6.** Dispersion stability by UV-Vis spectroscopy. The <sup>40</sup> dispersions are prepared with different dispersants (abscissa) by bath sonication, and measured immediately and 30 days after preparation. Relative absorption decrease is indicated. The absorbance (ordinate) is measured at  $\lambda$ =660 nm.

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**Figure S7:** Dispersant type versus graphene concentration and measured solvent surface tension. Surface tension was measured by pendant drop experiments with Attension Theta Optical Tensiometer. The dashed red area represents the optimal surface <sup>25</sup> tension range<sup>1</sup>.



**Figure S8.** (a) Pluronics parameters: total M<sub>w</sub> and number of <sup>30</sup> monomers in the hydrophilic (ethylene oxide-EO) and hydrophobic (propylene oxide - PO) blocks (n and m respectively, Figure S1F). (b) UV-Vis absorption of the dispersions prepared using Pluronics as dispersants in Tip-Bath-Tip procedure. The Pluronics (abscissa) are organized from left to <sup>35</sup> right with increasing number of PO units. Note that P-123 and F-127 have the same hydrophobic block's size but different





**Figure S9:** GS average length as measured by TEM. (a, b) TEM micrographs of small and large GS area. (c) GS average length <sup>55</sup> (square root of maximal × minimal lateral lengths) as a function of sonication energy (d) GS size distribution for all sonication energies. The stars in (a, b) denote the lacey carbon film.



**Figure S10:** Raman spectrum – 2D band Lorentzian line fit analysis. (a) The 2D band for a FLG has the denoted two <sup>5</sup> doublets. Since the Raman shift above 5 layers is hardly distinguishable from the bulk<sup>2</sup>, the high relative intensity of the lower frequency side of the FLG spectrum indicates that its thickness does not exceed 5 Layers. (b) 2D analysis of Graphite.<sup>2</sup>



**Figure S11.** AFM images of graphene sheets (GS) on SiO<sub>2</sub> wafer prepared from bath sonicated GS-TX-100 dispersion by spin coating. (a) GS with average length of 300nm (b) GS stack and (c) line profile (in b), indicating GS height of 1.3nm (lower right 15 part – full arrow) and 2.6 nm (center and upper left part – dashed

arrow).

<sup>35</sup> Table S1 - Summary of relevant parameters in GS dispersion in previous studies

Solvent (Surfactant)	Sonication time [hours] <sup>a</sup>			Final graphene	Ref.
	TS	BS	TS	concentration [mg mL <sup>-1</sup> ]	
Water (SDBS)		0.5		0.05	1
Water (SC)	1			0.09	3
Water (SC)		430		0.3	4
Water (TX-100)	0.83 b	1.33 b	0.83 b	0.7	This study
NMP		0.5		0.007	4
NMP		460		1.2	5
NMP		192		26-27	6
Chloro sulphonic acid				2	7
Pentafluor- obenzonitrile		1		0.1	8

<sup>a</sup> BS – bath sonication, TS –tip sonication.

<sup>b</sup> A combined procedure (tip-bath-tip), where both TS and BS are employed.

## References

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<sup>65</sup> 8.

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