

## ***Supporting information***

# **Increased Solubility and Fiber Spinning of Graphenide Dispersions Aided by Crown-Ethers**

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### **Materials**

The graphite powder (<20 micron, synthetic) was purchased from Sigma-Aldrich (batch #08017EH) and used as received. The HiPco SWCNTs (Product code: 195.1) used in the work were obtained from Rice University and purified as reported elsewhere.<sup>1</sup> Potassium metal was purchased from Sigma Aldrich. Naphthalene was purchased from Alfa Aesar. All of the reagents above were used as received. N-Methyl-2-pyrrolidone (NMP) was purchased from Sigma Aldrich and dried using molecular sieves followed by distillation under vacuum. 18-crown-6 was purchased from TCI and purified by recrystallization in dry acetonitrile.

### **Preparation of GIC**

The synthesis of GICs was performed using a modification of Penicaud group.<sup>2</sup> In a typical procedure, 60 mg of metallic potassium and 300 mg graphite powder were sealed in a heavy-walled glass tube under vacuum and heated for 48 hrs at 350 °C. The heavy-walled glass tube is then broken in a glovebox under nitrogen atmosphere and the GICs sealed in a 20 mL Wheaton vials for future use.

### **Potassium graphenide dispersions.**

For a typical preparation of a graphenide dispersion, 30-40 mg of GIC and a calculated amount of NMP were added into a 5 mL vial to get a desired initial concentrations, followed by stirring overnight. The mixtures were centrifuged at 9900 g for 30 min and the supernatants were analyzed using UV-Vis spectroscopy to determine its solubility based on the proportional constant reported elsewhere.<sup>2</sup>

For the dispersion of graphenide with crown ether, a crown ether solution of known concentration in NMP was added to the GIC. All the work was done in the glovebox under nitrogen atmosphere. For Raman spectra, soluble fraction (supernatant after centrifugation) and the insoluble fraction (precipitate after centrifugation), were quenched by methanol separately. The GIC solid was also quenched by methanol for comparison.

### **Mixed Graphenide/SWCNTDs dispersions.**

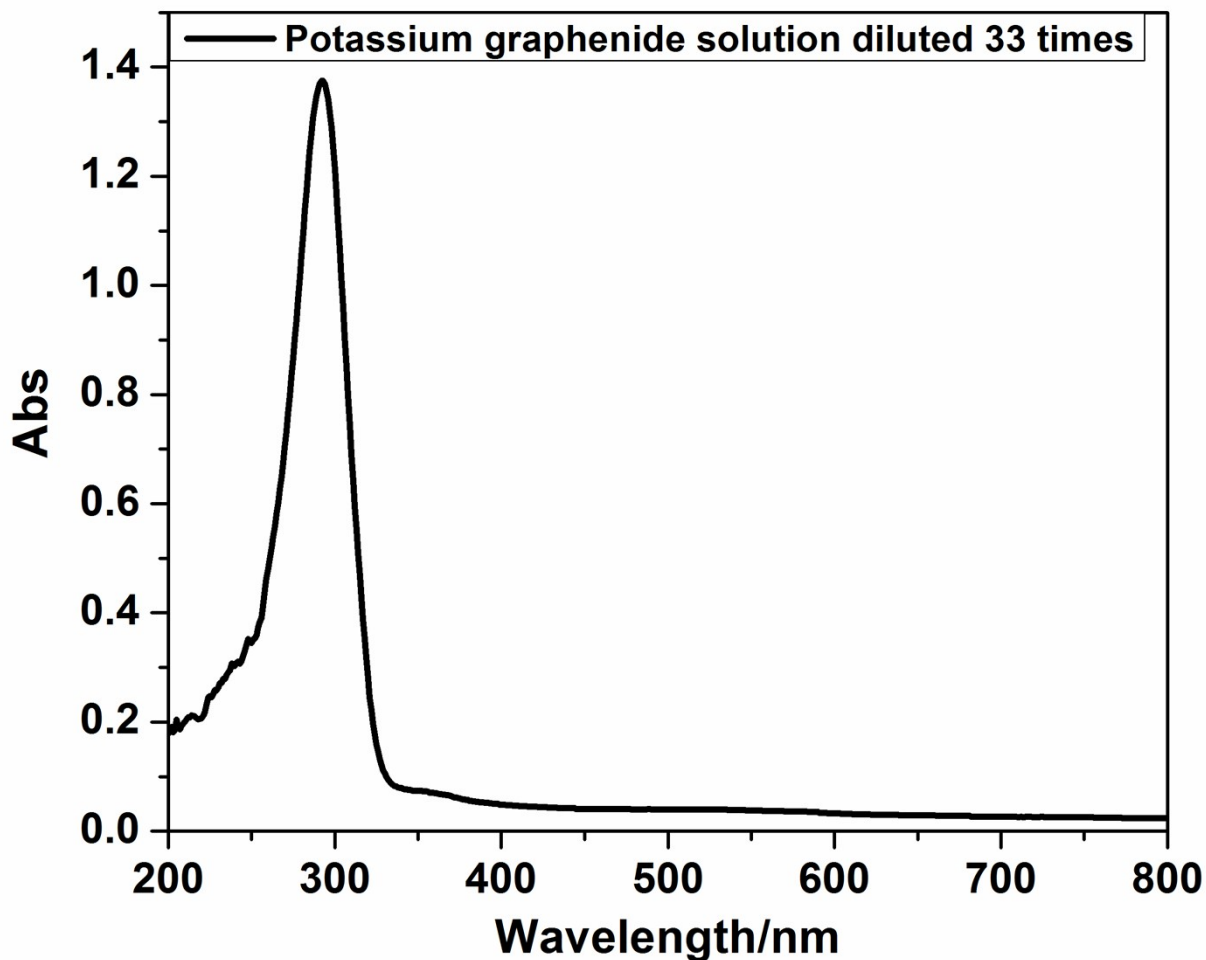
For the preparation of a typical Graphenide/SWCNTD dispersion with an initial concentration of 40 mg/mL SWCNTDs, 400 mg dry SWCNTDs solid (prepared as reported elsewhere<sup>3</sup>) was grinded with a mortar and mixed with 10 mL of 1.5 mg/mL Graphenide/8 mg/mL crown ether dispersion in NMP, with a calculated amount of extra 18-crown-6 to make the final concentration of crown ether to be 80 mg/mL. This is done to keep a constant proportion of SWCNTs to crown ether (20mg/mL of crown ether per every 10mg/mL of SWCNTs). The mixture was sealed in a 20 mL Wheaton glass vial and shaken for 1h at 1000 rpm on VWR mini shaker. Then the dispersion was further mixed for 1 h in a dual asymmetric centrifugation mixer (DAC 400.1 FVZ SpeedMixer) at 2350 rpm. The resulting dispersions were filtered through a 20  $\mu$ m mesh to remove large size aggregates. Similar procedures were adopted for the dispersions of 20 mg/mL and 30 mg/mL while maintaining the same ratio of SWCNTDs to crown ether.

### **Spinning of hybrid SWCNT/Graphene fibers.**

The spinning of SWCNT/graphene fibers was performed using the same set up used to spin SWCNTD fibers reported elsewhere.<sup>3</sup> SWCNT/graphene dispersions were loaded into a stainless steel injector and the dispersions were extruded through a 125  $\mu$ m tubing into coagulation solutions: water, 0.1 M HCl and 0.001 M  $\text{NaI}_3$  solutions. The resulting fibers were collected on a Teflon drum and immersed in water overnight to remove water-soluble impurities. Then the fibers were dried at 100 °C for 24 hrs.

### **Mechanical properties test of hybrid SWCNT/graphene fibers.**

The mechanical properties of SWCNT/graphene hybrid fibers were studied in a dynamic mechanical analysis system. The samples were tested with the assistance of 20 mm paper frames following a previous literature method.<sup>3, 4</sup> The diameters of the fibers were determined by scanning electron microscopy (SEM) images.



**Figure S1** UV spectrum of a typical graphene dispersion.

**Table S1** summary of SWCNT/Graphene hybrid fibers under different conditions

Initial concentration	20 mg/mL HiPco SWCNTD and 1.5 mg/mL graphenide with 40 mg/mL crown ether in DMSO			30 mg/mL HiPco SWCNTD and 1.5 mg/mL graphenide with 60 mg/mL crown ether in DMSO			40 mg/mL HiPco SWCNTD and 1.5 mg/mL graphenide with 80 mg/mL crown ether in DMSO		
Coagulation solution	water	0.1 M HCl	0.001 M NaI <sub>3</sub>	water	0.1 M HCl	0.001 M NaI <sub>3</sub>	water	0.1 M HCl	0.001 M NaI <sub>3</sub>
Diameter, $\mu\text{m}$	15 (3)	15 (3)	15 (3)	15 (2)	18(1)	14 (4)	18(3)	18(4)	18(3)
Tensile strength (MPa)	40(2)	32(5)	50(20)	100(10)	76(1)	100(10)	100(20)	110(10)	120(50)
Young's Modulus (GPa)	11(2)	7(2)	15(5)	23(3)	17(1)	25(2)	20(2)	22(2)	24(9)
Elongation (%)	0.4(0.2)	0.4(0.2)	0.4(0.2)	0.3(0.2)	0.09(0.02)	0.1(0.03)	0.5(0.2)	0.4(0.1)	0.4(0.1)
Conductivity (kS/m)	13(6)	9(2)	8(2)	30(10)	16(5)	50(30)	19(3)	16(2)	21(7)

## References

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