## **Supplementary Information**

# One-pot synthesis of conducting graphene polymer composites and their strain sensing application

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#### Synthesis of graphite oxide

Graphite oxide was prepared according to Modified Hummers method.  $^1$  SP-1 (Bay Carbon) graphite was used as the starting material. Briefly, graphite was grounded with NaCl and washed with deionized water followed by filtration using cellulose nitrate membrane. After drying, the filtrate was stirred with conc.  $H_2SO_4$  for 8 h. 6 g of KMnO<sub>4</sub> was gradually added while keeping the temperature less than  $20^{\circ}$ C. The mixture was stirred at 35 to 40  $^{\circ}$ C for 30 min and 65 to  $80^{\circ}$ C for 40 min. 92 ml water was added to the above solution and heated to  $\sim 100 \, ^{\circ}$ C. This was diluted by adding 280 ml of water followed by the addition of 30%  $H_2O_2$ . The mixture was washed followed by repeated centrifugation and filtration (first by 5% HCl and then with water). The final product was washed and dried in vacuum.

#### Synthesis of reduced graphite oxide in PVDF

Initially 10 mg of graphite oxide and 990 mg of PVDF was mixed with the help of mortar and pestle. PVDF was I whitish in colour and after adding GO also, composite looks like whitish, since GO was added in little and density of GO is high. This PVDF-GO was placed in petri dish and targeted towards focused solar electromagnetic radiation using 130 mm converging lens.

Temperature of the focused radiation was measured with type-K thermocouple. Care was taken with the converging lens such that temperature does not reach more than 250°C by adjusting the diameter of the converged beam. Similarly remaining compositions of graphene in PVDF has been prepared.

Solar graphene yield=~30%

PVDF (mg)	GO (mg)	Graphene in PVDF	wt.% of graphene in PVDF
		(mg)	(Gravimetric method)
990	10	3	0.3
970	30	9	0.9
950	50	15	1.5
930	70	21	2.2
900	100	30	3.2
850	150	45	5.0
800	200	60	6.9

Table S1: wt. % of reduced graphite oxide in polymer.

#### **Characterization techniques**

The XRD measurements were performed with a PANalytical X'Pert Pro X-ray diffractometer with nickel-filtered Cu K $\alpha$  radiation as the X-ray source. Raman spectroscopy was performed on Witec Raman microscope using green (532 nm) laser excitation, with excitation energy of 2.33 eV. Scans were taken on an extended range (500-3000 cm<sup>-1</sup>) for an exposure time of 60 s. Thermo gravimetric spectra were recorded with NETZSCH analyzer from ambient temperature to 800°C in  $O_2$  atmosphere with temperature heating of 10°C/min. Field emission scanning electron microscopy (FESEM, Quanta 3D) imaging was used to examine the morphology of the synthesized samples. Samples were mounted on a standard aluminium specimen stub using double sided conductive carbon tape. The microscope was operated at an accelerating voltage of 5-30 kV depending on different imaging purposes. EDX spectra was recorded with Li doped Silicon X-ray detector equipped with FESEM. High resolution micrographs were obtained with FEI Tecnai  $G_2$  transmission electron

microscope operated at 200 keV. Model 842.PE Newport optical thermopile based power meter was used to measure the power of radiation. Agilent thermocouple based temperature sensor was used to measure the temperature of the focused radiation.

# Fabrication of the specimen for strain sensing evolution

A nanocomposite film of reduced graphene-PVDF was attached to one side of the Aluminium (Al) specimen (Dimensions: 15 cm x 3 cm x 0.3 cm) using high strength epoxy to make perfect bonding between Al and the nanocomposite film. On the other side of specimen, a conventional metallic strain gauge was adhered using glue. Resistance of the composite film was measured with Keithley source meter 2400 and data was continuously acquired using Lab VIEW programme while specimen undergoes tensile test. Al specimen was subjected to ramping loads, tensile and compressive loads on materials testing system (MAKRON, 25 kN) which is servo hydraulic test machine. The corresponding strain in the composite film was measured by conventional metallic strain gauge.

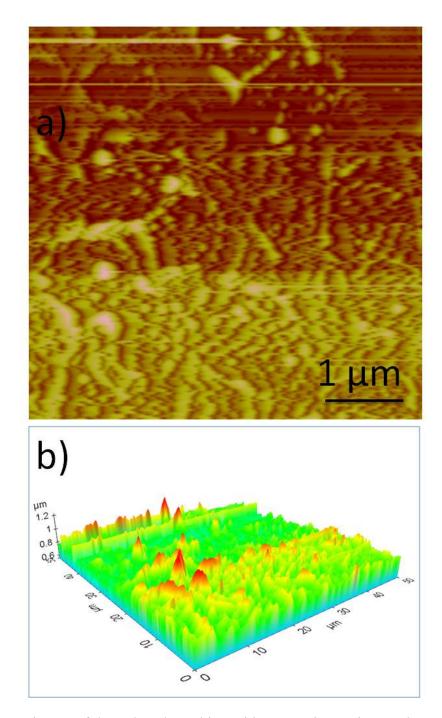


Figure S1: a) AFM image of the reduced graphite oxide-PVDF in tapping mode and b) its 3D image.

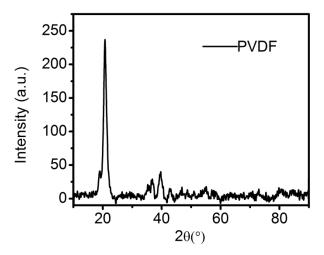


Figure S2: XRD of pure PVDF after solar irradiation

The XRD of pure PVDF after melting with focused solar radiation is shown in above figure S2. It exhibits both phases of alpha and beta. When graphite oxide is added to the PVDF and performed photo chemical reduction, most preferable beta phase is forming due to the less binding energy of beta phase to the surface attachment of reduced graphene.

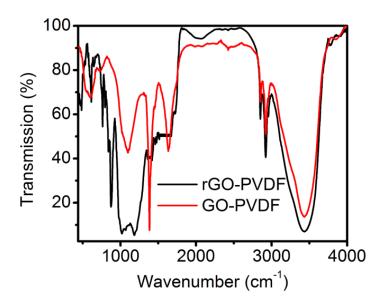


Figure S3: FTIR of graphite oxide and reduced graphite oxide

The synthesis of graphene by reduction of graphite oxide using focused electromagnetic radiation has been reported by our group. Figure S3 shows the FTIR of neat GO and reduced graphite oxide. Graphite oxide (GO) identified the presence of following functional groups. O-H stretching vibrations (3432 cm<sup>-1</sup>), CH<sub>2</sub> asymmetric and symmetric stretching vibrations (2924 and 2852 cm<sup>-1</sup>), C=O stretching vibrations (1725 cm<sup>-1</sup>), C=C from unoxidized sp<sup>2</sup> bonds (1627 cm<sup>-1</sup>), O-H bending deformation (1408 cm<sup>-1</sup>) and C-O vibrations (1049 cm<sup>-1</sup>). This confirms the presence of hydroxyl, epoxide and carbonyl functional groups in GO as shown in the above figure. On the other hand, the spectrum of graphene exhibits a broad O-H band (3436 cm<sup>-1</sup>) which can be due to adsorbed moisture. The absence of any other oxygen containing functional groups in graphene clearly validates the exfoliation process. Figure S4 also shows the TGA of graphite oxide and reduced graphite oxide. Thermo gravimetric analysis (TGA) of graphite shows GO is thermally unstable and starts to lose mass upon heating below 100 °C due to moisture and water, but severe mass reduction occurs at 190°C probably due to pyrolysis of the labile oxygen-containing functional groups, yielding CO, CO<sub>2</sub>, and steam. Hence, the thermal decomposition of GO can be accompanied by a vigorous release of gas, resulting in a rapid thermal expansion of the material. This is very clear from the video, where upon irradiation with sunlight, rapid generation of gases is visible. Irradiation imparts thermal stability to the reduced GO with the removal of thermally labile oxygen functional groups. Apart from a 6 wt % loss below 200°C, which can be due to the release of remaining oxygen functionalities, no significant mass loss is detected when solar graphene is heated up to 600°C.

Completely exfoliated graphene sheets can be observed in the polymer matrix and it can seen in AFM image as shown in figure 1.

#### **Electrical conductivity**

Two probe method was employed for measuring electrical conductivity of the reduced graphite oxide (rGO)-PVDF with higher resistivity whereas for reduced graphene composites with semiconducting or conducting in nature, collinear four probe technique was used in order to avoid contact resistance, lead resistance etc. Positive constant current was applied to the first and fourth terminal and corresponding voltage was measured between second and third terminals. Similarly negative constant current was applied and corresponding voltage was measured. Positive and negative currents were applied in order to avoid voltage generation due to thermoelectric effects. All input and measured parameters were substituted in equations 1, 2 and 3 for calculating conductivities of the samples insulating, semiconducting and conducting

samples. We also have done transverse electrical measurements on these reduced graphite oxide-PVDF thin films.

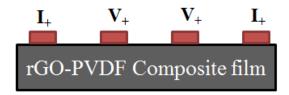


Fig. S4: Representation of in-plane conductivity measurements using four probe set up

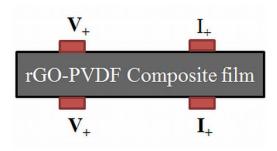


Fig.S5: Representation of transverse conductivity measurements using four probe set up

Two probe technique, electrical resistivity ( $\Omega$ -m)

$$R = \frac{\rho l}{A} - \dots (1)$$

where R is the measured resistance between the two probes  $(\Omega)$ 

1 is the probe separation (m)

and A is the area of current distribution (m<sup>2</sup>)

Four point collinear probe technique

$$\rho = \frac{\pi t}{\ln 2} \frac{(V_{+} - V_{-})}{2I} k - \dots (2)$$

where  $\rho$  is electrical resistivity ( $\Omega$ -m)

t is the thickness of the sample (m)

I is the applied current (A)

 $k = k_1 k_2$  is the correction factor due to finite size and dimensions

V<sub>+</sub> is the measured voltage for applied positive current (V)

V<sub>2</sub> is the measured voltage for applied negative current (V)

Reciprocal of resistivity is known as conductivity ( $\sigma$ )

$$\sigma = \frac{1}{\rho} \text{ (S/m)} - \cdots (3)$$

# **Energy dispersive analysis of X-rays (EDAX)**

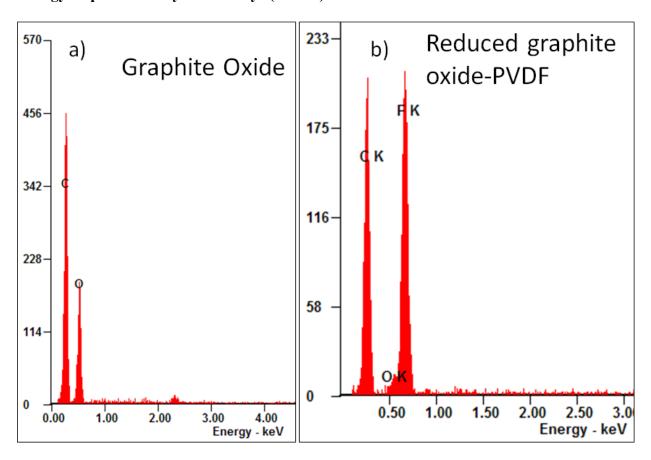


Fig. S6: Energy dispersive X-ray spectra of graphite oxide and reduced graphite oxide-PVDF.

# References

1. W. S. Hummers and R. E. Offeman, *Journal of the American Chemical Society*, 1958, **80**, 1339-1339.