Supplementary Data

Low temperature processed graphene thin film transparent electrodes for supercapacitor applications.

Shriniwas Yadav [a,b], Inderpreet Kaur* [a,b]

[a] Central Scientific Instruments Organisation (CSIR-CSIO), Sector-30C, Chandigarh, 160030, India.
[b] Academy of Scientific and Innovative Research, (AcSIR) CSIR-CSIO, Sector-30C, Chandigarh, 160030, India.

1. **Figure of merit (σ_{dc}/σ_{opt}) values for graphene based films made by different methods are tabulated below:-**

Electrode Material	Preparation Method	Transmittance (%T at λ=550 nm)	Sheet Resistance (KΩ/sq)	FoM (σ_{dc}/σ_{opt})	References
G-COOH /NH_2-G	Spin coating	84-94	0.43-18.7	4.8	(in this work)
Graphene/ MWCNT	Vacuum filtration	77-87	0.13-1.2	9.84	[1] Yadav S. et al., Thin solid films 2015.
Reduced Graphene Oxide (rGO)	Drop casting	70	0.2	4.81	[2] A. Nekahi et al. Applied Surface Science 2014.
CVD grown Graphene	Etching Ni followed by scooping	87	0.60	4.3	[3] M. Choe et al. Org. Electron. 2010.
rGO	Hydrazine reduction & thermal annealing at 700 ° C	69	17.9	0.05	[4] Y. Xu et al. Carbon 2010.
rGO/CNT	Hydrazine reduction	86	0.24	10	[5] V.C. Tung et al. Nano Lett. 2009.

Table 1. Figure of merit (σ_{dc}/σ_{opt}) values for graphene based films made by different methods.

2. FTIR characterization of Graphene-COOH and Graphene-NH_2 used in our work:

For carboxylation, Graphene (15mg) was treated with H_2SO_4: HNO_3 in 1:3 ratio by ultrasonicating for 15 minutes in ice bath. After ultrasonicating, Deionized (DI) water was added into the mixture and left for 30 minutes at room temperature. Then the solution was filtered and washed five times with 18 MΩ DI water through the hydrophilic filtration membrane (0.22 µm, pore size). Filtered graphene was re-dispersed in DI water and the solution was centrifuged 5 times at 2000 rpm for two minutes to remove all the insoluble particles. Supernatant solution was diluted to 1.5 mg/ml and used for further experimentation. The FTIR spectrum of G-COOH (Figure S1) demonstrates the presence of C–O (peak at 1090cm^{-1}), C–OH (peak at 1380 cm^{-1}), C=O in carboxylic acid and carbonyl moieties (peak at 1620 and 1720 cm^{-1}) and the H-bonded associated OH (broad band around 3430 cm^{-1}).[6]

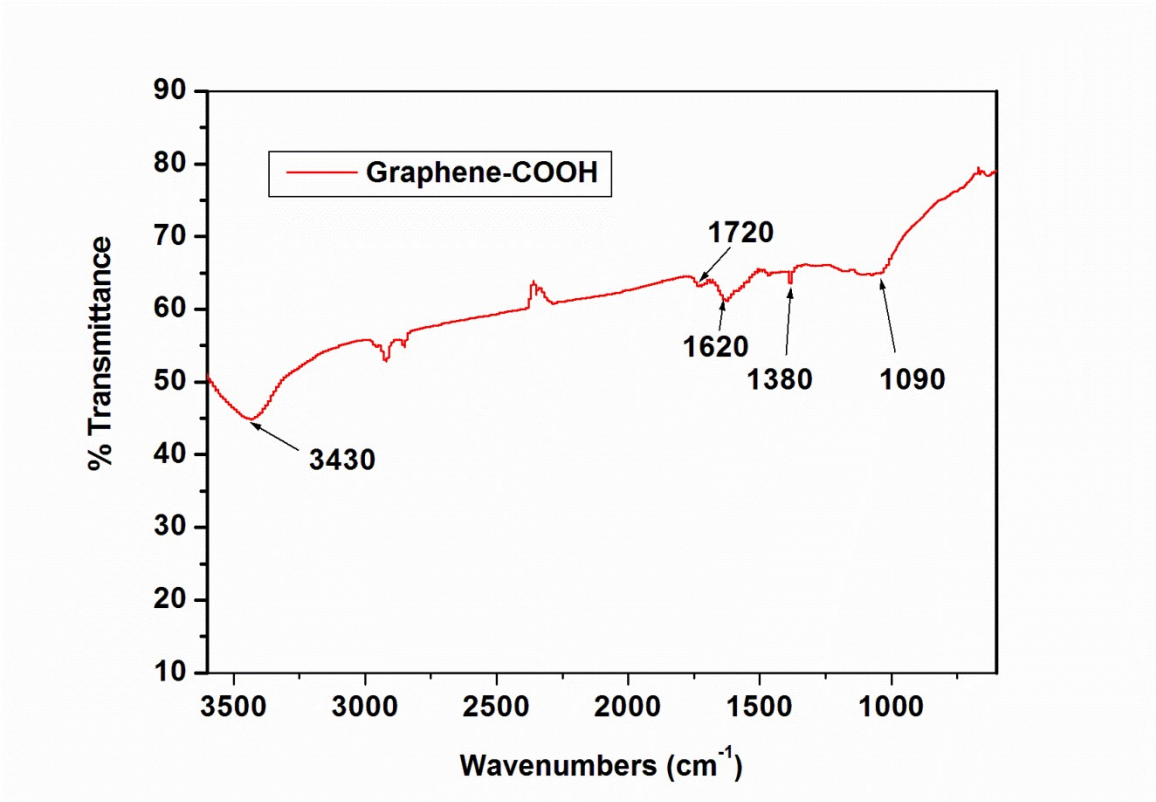

Figure S1:- FTIR spectra of Graphene-COOH.

To prepare amide functionalized graphene, carboxylated graphene was used. The suspension of carboxylated Graphene (100ml, 0.15 mg/ml) was reacted with ethylenediamine (3.0 ml) under stirring for 5 h in the presence of 300 mg of EDC and 0.5 ml liquor amonia. The resulting suspension was sonicated for 30 minutes prior to vacuum filtration. Filtered Graphene was re-dispersed in DI water and the solution was centrifuged 5 times at 2000rpm for two minutes to remove all the insoluble particles. Supernatant solution was diluted to 1.5 mg/ml and used for further experimentation. FTIR spectra of Graphene-NH_2 (figure S4) shows peak at 1640 and 1020 cm^{-1}, corresponding to N-H and C-N in-plane stretching.[7] This reflects successful introduction of nitrogen group. A low intensity band at 3400-3500cm^{-1} remains, which may be attributed to the N-H stretching of the amine groups.[7]

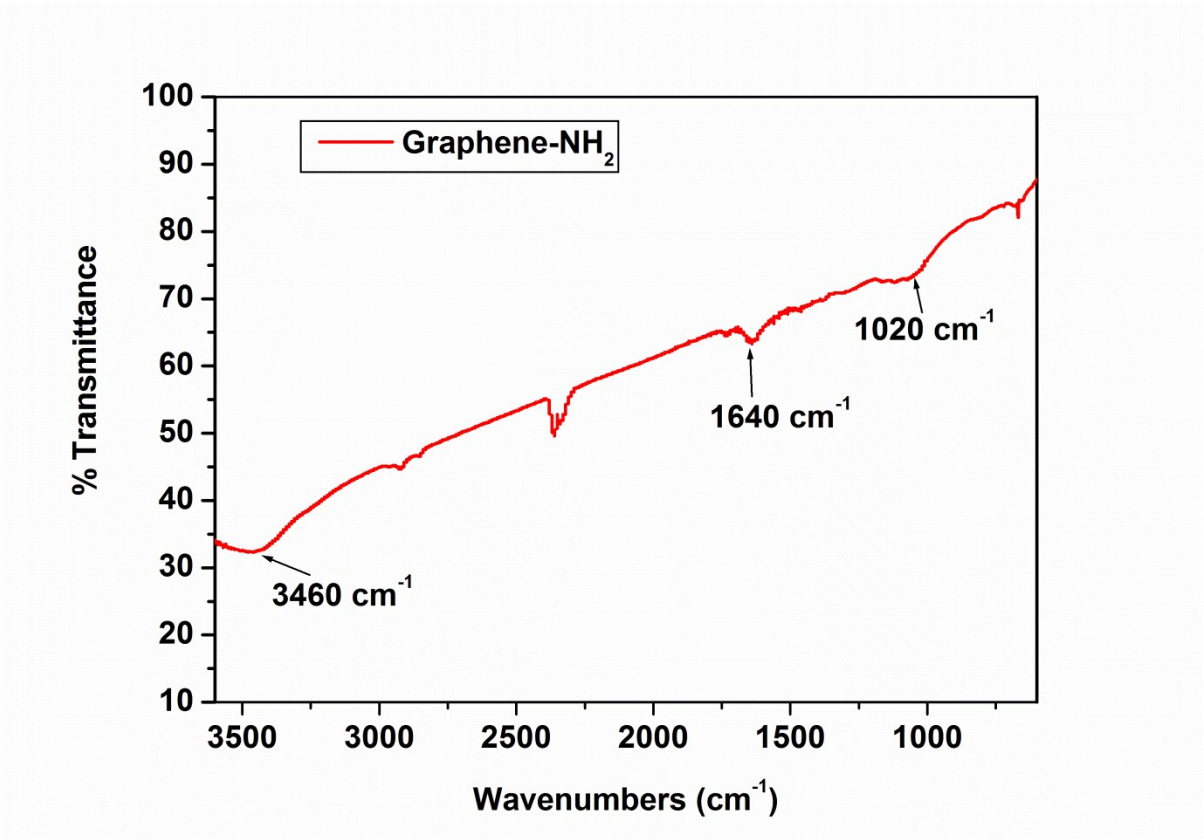

Figure S2:- FTIR spectra of Graphene-NH_2.

3. Graphene weight measurement:

The amount of graphene in G-COOH/NH_2-G thin films was calculated by Lambert-Beer law following previously reported procedure.[8] It was assumed that the extinction coefficient of carboxylic acid and amide functionalized graphene is same in dispersion and thin film. A series of graphene dispersions of known concentrations in DI water were prepared and measured for absorbance at 550 nm using UV-Vis spectrophotometer. Absorbance (A) vs. concentration (C) curve was plotted as shown below.

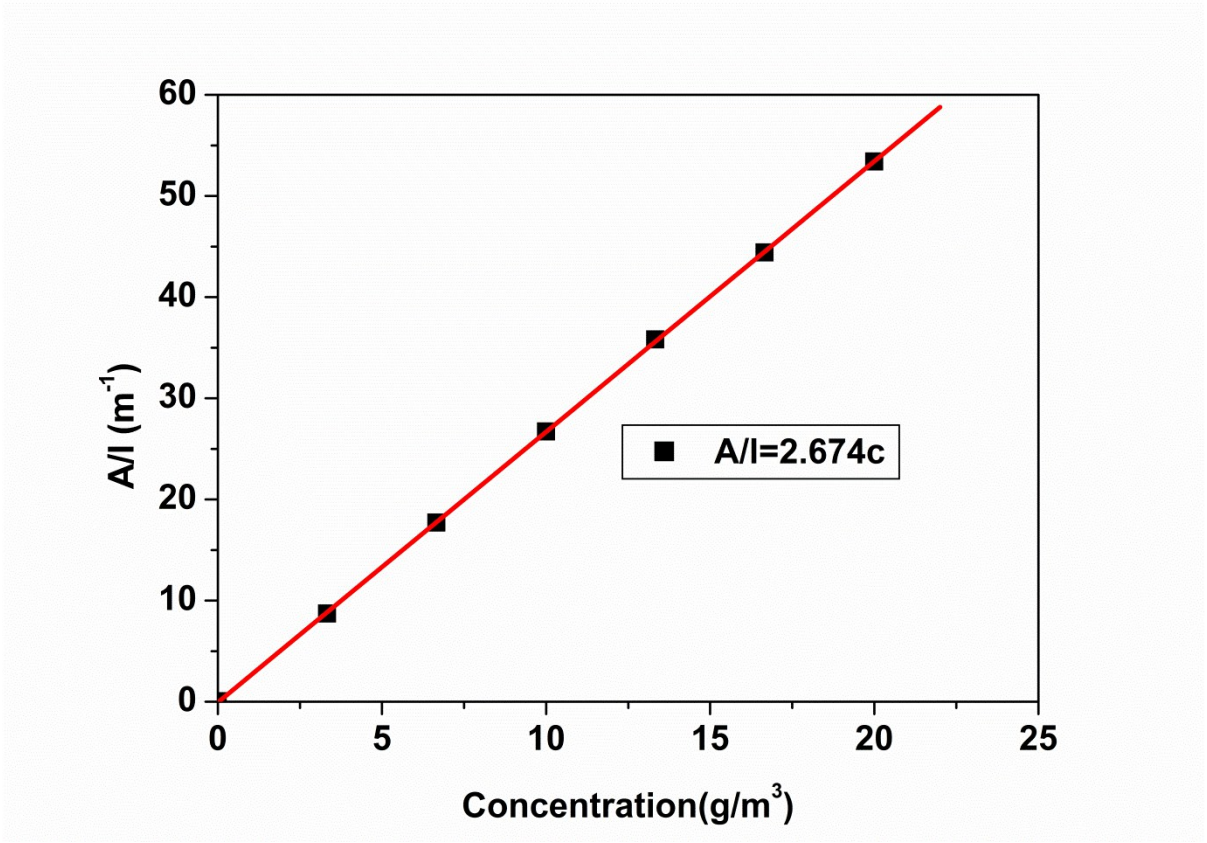

Figure S3: The relationship between the absorbance at wavelength of 550nm and concentration of G-COOH in DI water.

Here l refers to the thickness of sample cell. The abscissa corresponds to the concentration of G-COOH in dispersion. The amount of graphene was calculated by using following equation based on the Lambert-Beer law.

$$m = C.S.b = \frac{A.S}{2.674}$$

Where S is the size of the film (m^2), b is the film thickness and A is the absorbance at a wavelength of 550nm.

Using above relation amount of graphene in thin film fabricated at various spin speeds are calculated and tabulated below.

S. No.	Spin speed (rpm)	Absorbance	Size (m^2)	Weight of graphene (μg)
1.	1000	0.159	0.0001	5.94
2.	1200	0.146	0.0001	5.45
3.	1400	0.122	0.0001	4.45
4.	1600	0.058	0.0001	2.16

Table 2: Calculated mass of graphene in different thin films fabricated at spin speed from 1000 to 1600rpm.

4. EIS measure of Graphene thin film over glass substrate:

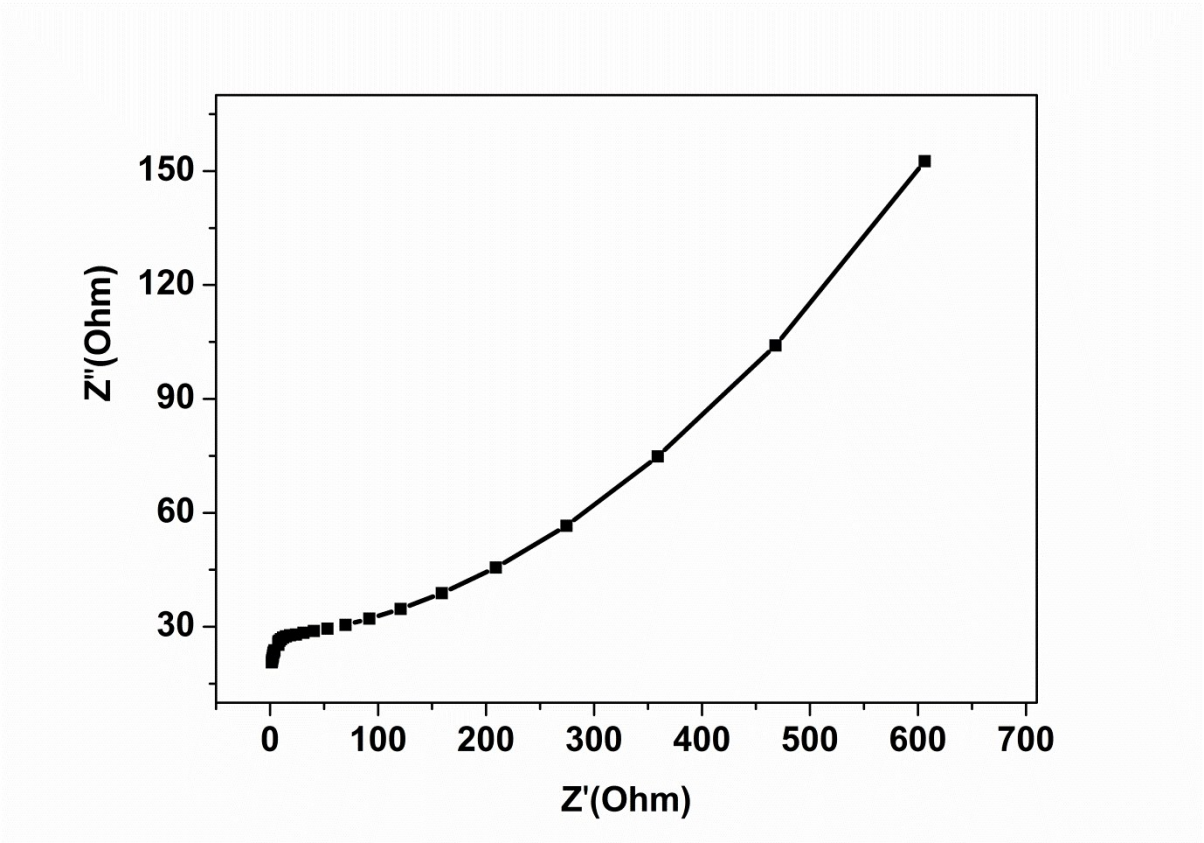

Figure S4:- Nyquist plot for the graphene film fabricated at spin speed of 1600rpm in the frequency range from 1MHz to 100 mHz at an ac amplitude of 10mV.

Nyquist plot of the thin film fabricated at a spin speed of 1600 rpm is shown in figure S4. The imaginary component (Z″) of the impedance is plotted versus the real component (Z′). The frequency response of the film from 1MHz (lower left portion) to 100 mHz is shown by

curve. The incomplete arc of frequency response toward higher frequency related to electronic resistance of the thin film electrode.

References

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