Supplementary Information

Direct Synthesis of Large Area Graphene on Insulating Substrates at Low Temperature using Microwave Plasma CVD

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Table. S1 Sheet resistance and transmittance of graphene synthesized at different microwave powers

Table S1 explains the dependence of sheet resistance and transmittance on microwave power. Experiments were performed at microwave power from 700W to 1300W and it can be easily observed from table 3 that transmittance decreases as we increase power. Although, sheet resistance decreases but the simultaneous decrease in transmittance also occurs.

Figure. S1 Dependence of sheet resistance and transmittance on deposition time

Therefore, microwave power 1000W was chosen to perform the further experiment as transmittance and sheet resistance of graphene directly grown on glass and quartz at this power was acceptable. After confirming the acceptable microwave plasma power optimum time for deposition was obtained by performing various experiments at different time periods as shown in figure S1. It shows, lesser the time of deposition, higher the

sheet resistance and transmittance and *vice versa*. Deposition time between 2-4 minutes form graphene with sheet resistance ranging from 1300 Ω/\Box to 2400 Ω/\Box and transmittance varied from 60% to 80%. In order to obtain graphene with 80% and more with sheet resistance as low as possible, we concentrated on performing more and more experiments in 2 to 4 minutes of deposition time using microwave plasma CVD.

Figure. S2 a) 3-Dimentional, b) 2-Dimentional optical image of glass surface after graphene deposition without CO2, c) Raman spectrum of deposited graphene (with inset showing place of Raman measurement)

Figure S2 a) shows a 3-dimensional optical image of graphene on glass without $CO₂$ use,

indicating the formation of so vertical structures. Figure S2 b) shows a typical optical image of a glass surface after graphene deposition.

Figure. S3 Optical transmittance of graphene deposited on quartz

Optical transmittance characterization of the graphene deposited glass and quartz surfaces were carried out with UV-Vis-NIR on a JASCOV570 spectrophotometer. Fig. S3 shows graphene with transmittance 70-85% in visible and near infra-red regions. Thus formed graphene has acceptable transmittance in the visible region and better transmittance as compared to Indium Tin Oxide in near infra-red regions.

Also, Table S2 shows that ozone treatment has helped in the removal of surface contaminations of graphene, as there is a decrease in carbon percentage causing transmittance to increase by 1%. Whereas, an increase in oxygen and silicon atomic concentrations might be due to the removal of unwanted impurities causing x-rays to detect $SiO₂$ surface beneath graphene comparatively more easily. Figure S4 also confirms the reduction of impurities of graphene samples after ozone treatment as there is a decrease in Id/Ig ratio.

	C1s (%)	N1s(%)	$O1s$ (%)	Si2p(%)
As-grown graphene	97.2	0.0	1.5	1.3
O3 treated graphene	94.7	0.0	3.5	1.8

Table. S2 Surface composition of graphene grown quartz surface

Figure. S4 Raman spectra of quartz surface before and after ozone treatment

Figure. S5 a) AFM image of the Silicon surface after graphene deposition for 2 minutes, b) Line profile at the highlighted part on the surface

	Carrier Mobility $\rm (cm^2/$	Sheet Carrier	Sheet
Tec	$V.s$)	Concentration (/cm^2)	Resistance
			(Ω/\Box)
Before Ozone treatment	9.64	$2.89x10^{14}$	2240
After Ozone treatment	97.5	$5.09x10^{13}$	1259
After Ozone treatment	54.75	$5.09x10^{13}$	Assuming no effect of
After Ozone treatment	97.5	$2.86x10^{13}$	Ozone on R_s

Table. S3 Effect of ozone treatment on carrier mobility and carrier concentration of graphene

Lake Shore 8400 series Hall measurement instrument was used for sheet carrier and mobility of the samples. This instrument contains the Van der Pauw measurement technique and the formula used for sheet resistance (Rs) calculation,

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R_s = \frac{1}{qn_s\mu_m}
$$

Where,

Rs= Sheet resistance

q= Charge on the carriers

ns= Sheet carrier concentration

 μ_m = Mobility of the majority carriers

After cross checking the calculations it was noticed that if ozone treatment had no impact on graphene sheet resistance then the product of n_s , μ_m before and after ozone treatment should remain the same, and in such case Hall measurement after ozone treatment should yield lower value of carrier mobility (54.75 cm²/ V.s) corresponding to 5.09 x 10^{13} cm⁻² carriers as shown table S3. However, mobility was found to be $97.5 \text{ cm}^2/\text{V}$. which indicates that there is an effect of ozone treatment on graphene and additional 42.75 cm^2 / V.s of mobility can be responsible for sheet resistance reduction to 1259 Ω/\Box from 2240 Ω/\Box

Similarly, in the case of carrier concentration, Hall measurement after ozone treatment should yield lower value of carrier concentration $(2.86 \times 10^{13} \text{ cm}^{-2})$ corresponding to 97.5 cm²/ V.s carrier mobility as shown table S3. However, carrier concentrations was found to be 5.09 x 10^{13} cm⁻² indicating ozone treatment helps in increasing p-doing of graphene, as there is an addition in majority carriers by an amount of 2.23 x 10^{13} cm⁻² in the calculated value of 2.86 x 10^{13} cm⁻². As reported by Jandhyala *et al.*, physisorbed ozone can be responsible for the upshift of the Dirac point, leading to p-type doping of mechanically exfoliated graphene. With the increase in p-type doping, improved electrical conductivity is expected.

Above mentioned, Hall data and calculations show that carrier concentration and mobility increase after ozone treatment by 43.8% and 56.2%, respectively, from the calculated value in case when no effect of ozone on graphene is assumed.