## Electrophoretic separation and deposition of metal -graphene

## nanocomposites and their application as electrodes in solar cells

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The Raman spectra recorded for the D band and G band for all of the materials under investigation (GO, rGO, PtrGO and AurGO) are presented below. The materials were prepared via a hydride reduction of GO in the presence of gold and platinum salts, as described in the methods section below. The rGO was prepared through a reduction with NaBH<sub>4</sub>, without the presence of any metal salt. A highlight of the Raman G band for the materials is shown in Fig. **SI2** and the corresponding G band positions are represented in Table 1. It can be observed from Table 1 that the G band position for graphite and GO shifts significantly upon oxidation (1578 .80 cm<sup>-1</sup> to 1583.88cm<sup>-1</sup>). Upon reduction of GO with NaBH<sub>4</sub> the G band position shits back to the position of graphite (1578.25 cm<sup>-1</sup>) indicating an increased level of graphitisation in the material. This shift is also observed for AurGO and PtrGO, but not to the same extent. This is most likely due to NaBH<sub>4</sub> acting as a co-reducing agent for both the metal salts and the GO simultaneously. Hence, a less significant blue shift is observed.



Fig. SI 1 Raman spectra of D and G band for GO, rGO, AurGO and PtrGO.

The ratio of  $I_D/I_G$  (D band intensity/G band intensity) is often used as a gauge for measuring the extent of disorder caused by defects in carbon nanomaterials. Generally it is assumed that a higher  $I_D/I_G$ indicates greater level of disorder in these materials however some results in the literature do not support this argument. The prominent increase in the D band intensity is related to a reduction in size of the in-plane sp<sup>2</sup> domains. The reduction of GO to rGO does not significantly alter the  $I_D/I_G$  ratio (0.971 and 0.976 respectively). After deposition of the metallic nanoparticles  $I_D/I_G$  increases slightly to 1.002 for AurGO and 1.040 for PtrGO. The increase in the  $I_D/I_G$  ration has been linked to the creation of new graphitic domains, which are smaller in size than that present in GO but more numerous in number.

The 2D band in the Raman spectrum of graphene oxide is seen to broaden and decrease in intensity, as seen in Fig. **SI 2**. This is a result of the increase presence of defects in the material. The return of the 2D band after the reduction with NaBH<sub>4</sub> indicates that it can successfully partially restore the sp<sup>2</sup> nature of the lattice.<sup>1,2</sup>



**Fig. SI 2** Left: shift in position of Raman G band for graphite, rGO, AurGO, PtrGO and GO and right: Raman spectra of 2D band of GO, rGO, AurGO and PtrGO.

Material	G band (cm <sup>-1</sup> )
Graphite	1578.80
rGO	1578.25
AurGO	1578.25
PtrGO	1581.45
GO	1583.86

**SI Table 1.** Shift in position of Raman G band for graphite, rGO, AurGO, PtrGO and GO

XRD patterns were recorded for GO, rGO, PtrGO and AurGO and are presented in the **SI 3**. The XRD pattern for GO, which is composed of 2 distinct peaks located at  $2\theta = 10.89^{\circ}$  and 26.68°, the first peak is due to the diffraction cause by GO while the second peak is attributed to graphite (Fig. **SI 3a**). This peak most likely arises from areas of un-oxidised regions in the sample. Upon reduction of GO to rGO the peak located at  $10.89^{\circ}$  disappears completely from the diffraction pattern, thus indicating the reduction of the functional groups on the GO surface (Fig. **SI 3b**). The peak associated with graphite diffraction peaks associated with both Au and Pt to appear. Figure 9c displays the diffraction pattern recorded for PtrGO. Pt peaks at  $2\theta = 39.3^{\circ}$ ,  $46.2^{\circ}$ ,  $67.9^{\circ}$ , and  $81.4^{\circ}$  can be assigned to the (111), (200), (220) and (311) crystalline planes of Pt. This indicates that the deposition of Pt onto GO allows for the formation of NPs that are composed of pure crystalline Pt. Figure 9d shows the diffraction pattern recorded for AurGO. The diffraction peaks are similar to that of pure Au, with peaks at  $2\theta = 33.26^{\circ}$ ,  $44.60^{\circ}$ ,  $67.67^{\circ}$ ,  $77.54^{\circ}$  and  $82.35^{\circ}$  assigned to (111), (200), (311) and (222) crystalline planes of gold



Fig. SI 3 XRD patterns recorded for GO, rGO, PtrGO and AurGO.





Fig. SI 4 CVs recorded for AurGO (top) and GO (bottom) electrodes



Fig. SI 5 J-V curves recorded for DSSCs employing AurGO CEs after multiple deposition cycles



Fig. SI 6 J-V curves recorded for DSSCs employing PtrGO CEs after multiple deposition cycles

## References

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