Supporting Information for
“Highly conductive graphene sheets through low-temperature reduction of partially oxidized graphene”

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S1. AFM images
For AFM studies, single-layer rPOG films were prepared on SiO\textsubscript{2}/Si substrates through L-B assembly as mentioned in the manuscript. The samples were annealed at 250 °C in N\textsubscript{2} atmosphere prior to imaging. The measurements were conducted on Agilent 5500 AFM in ambient condition. Imaging was carried out in tapping mode with standard NCH (non-contact, high resonance frequency) cantilevers with spring constant of ~40N/m and resonant frequency of ~300 kHz. As shown in Figure S1, the substrate surface is partially covered by rPOG sheets. The individual sheets exhibit unique surface features due to wrinkles. The height profiles clearly show that most sheets have uniform thickness of about 0.9 ~ 1.2 nm, which corresponds to the thickness of typical single-layer chemically derived graphene sheets\textsuperscript{51}, indicating that the POG sheets are predominantly single-layered. Overlapping sheets can also be readily seen in these films. The thickness of these regions is roughly a multiple of 1 nm. rPOG films prepared from a single deposition after L-B assembly has non-uniform thickness but the average thickness is roughly 1 nm.
S2. Optical image

It is well established that under optical microscope graphene exhibits layer contrast on silicon substrates with 300 nm thermal oxide due to interference effects\(^{S2}\). To check for thickness uniformity, single-layer POG films were prepared on SiO\(_2\) (300 nm)/Si substrates as described in the manuscript and observed under optical microscope. Figure S2 clearly shows that majority of the sheets exhibit the same contrast, indicating that these sheets have the same thickness. Some sheets which exhibit darker and varying contrast due to larger thicknesses are also visible in minor quantity. Based on the agreement with the AFM results, it can be concluded that a majority of the flakes is single-layered.

Figure S1 – AFM images of monolayer rPOG film and corresponding height profiles.
S3. Transparent and conductive properties

We have evaluated the transparency and conductivity of the rPOG films. Figure S3 shows the transmittance ($T$) and sheet resistance ($R_s$) of rPOG in comparison with those of GO reduced in different conditions. The data for rGO were obtained from Ref. S3. It can be clearly seen that after mild annealing, rPOG exhibits significantly improved transparency and conductivity compared to rGO. However, high temperature annealing does not significantly improve the conductivity of rPOG films further and leads to properties comparable to those of rGO reduced at similar conditions. This may be explained if the film conductivity is dominated by junction resistance between individual sheets rather than the conductivity of individual sheets in this regime of reduction. Since the nature of the sheet junctions is likely the same for rPOG and rGO, only minor differences are observed between these samples. The size of individual flakes is expected to have more dominant effects on the overall conductivity of the films in this reduction regime S4. Further, other effects such as charged impurities may also have some effect on the overall conductivity of these films.

Figure S3 – Transmittance ($T$) at 550 nm vs sheet resistance ($R_s$) for rPOG and rGO thin
films with various thicknesses and reduction conditions.

S4. References

S5. Complete list of authors and the full citation for Ref 6: