Electronic Supplementary Material (ESI) for Nanoscale.

Supplementary Information

Voltage-reduced low-defect graphene oxide: a high conductivity, near-zero temperature coefficient of resistance material

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1. Oxo-G Sheet Size and Film Thickness

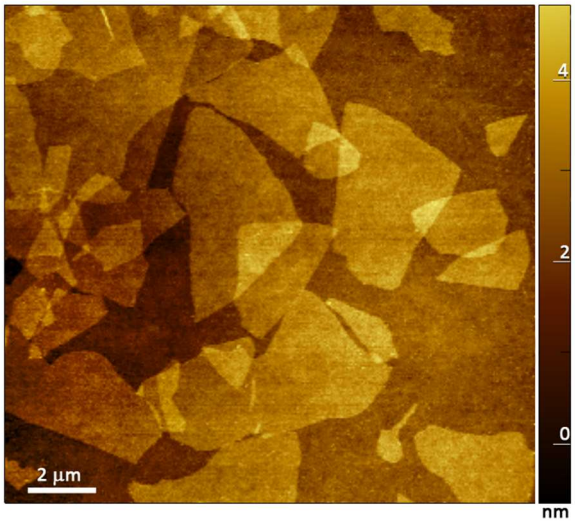

Figure S-1. Tapping mode atomic force microscope topography of oxo-G sheets on a glass substrate. The average sheet area based on measurements of 50 sheets is 10.7 μm^2 with a standard deviation of 2.6 μm^2.

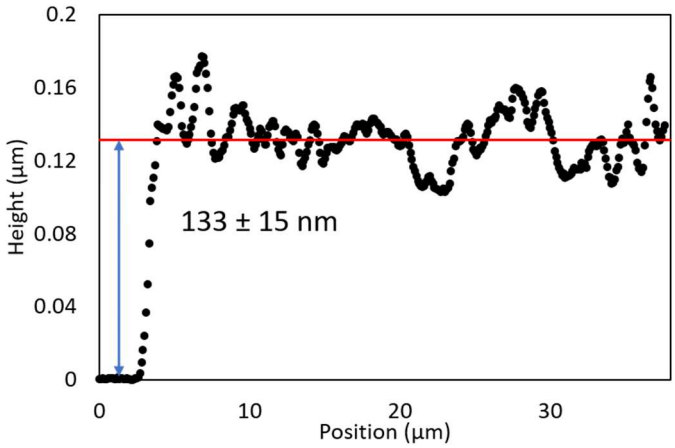

Figure S-2. Atomic force microcsope line profile, averaged over 50 scan lines, showing the height difference between a bare glass substrate and the surface of a V-roxo-G film. The glass region was exposed by scratching the V-roxo-G film with a sharp probe. The film thickness was determined from the average height difference to be 133 ± 15 nm in this case. Local V-roxo-G film thickness within each electrode gap was measured at two or more locations to confirm that the thickness in a given gap is constant to within about 10%, and to ensure an accurate representation of film thickness when determining film conductivity. The conductivity was found to be the same for all measured V-roxo-G film thicknesses (ranging between 40 and 150 nm).

2. Oxo-G Reduction

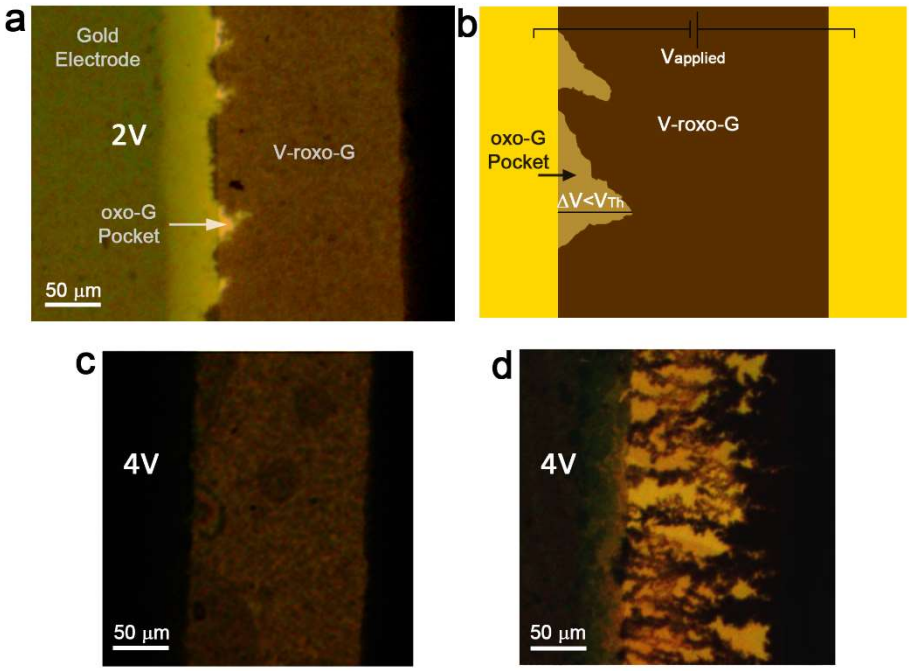

Figure S-3. a) Voltage reduction of oxo-G at 2 V for 5 hours at 70% RH showing nearly complete reduction, but also small pockets of unreduced oxo-G. b) Schematic depicting the proposed mechanism for pocket formation: before the V-roxo-G reaches the positive electrode, the potential difference between the V-roxo-G and the positive electrode is sufficient to maintain reduction. However, once V-roxo-G reaches the positive electrode, nearby V-roxo-G is at a potential that is close to that of the positive electrode, halting reduction. A threshold voltage, V_{Th}, of about 1.5 V is required for reduction to proceed. As shown in (c), reduction can be completed by applying a second reduction step at a higher voltage, 4 V for 1 hour. d) If 4 V is applied from the start, branching V-roxo-G filaments are formed that leave large unreduced pockets of oxo-G. Near-complete reduction can be achieved in tens of minutes by using an intermediate voltage; however, to ensure accurate conductivity measurements, complete reduction was achieved by using a slower 2-step process. The reduction kinetics depend on voltage, film thickness, and humidity. Since a relatively low voltage (low rate) was employed, relatively thick films (40 – 150 nm) and high humidity (70% RH) were used to accelerate the reduction process.

3. Variable Temperature Measurements

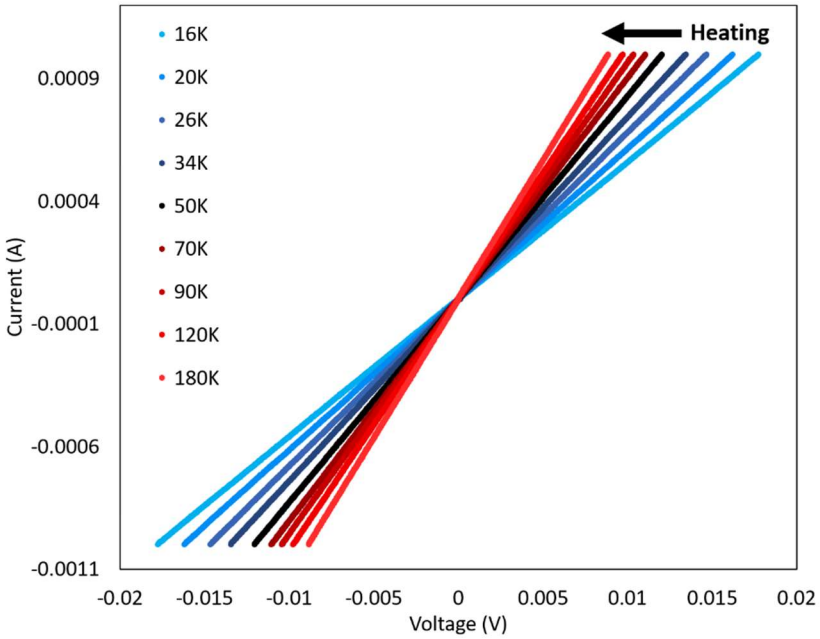

Figure S-4. Current-voltage characteristics of V-roxo-G as temperature is varied between 16 and 180 K, showing ohmic behavior. Ohmic behavior was observed over the entire 16 – 373 K range. Temperatures above 180 K are not shown due to the minimal temperature dependence leading to overlap of the I-V curves.