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Investigation of the high-field transport, Joule-heating-driven conductivity improvement and low-field resistivity behaviour in lightly-reduced free-standing graphene oxide papers

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Free-standing reduced graphene oxide (rGO) has been gaining popularity for its use in supercapacitors and battery applications due its facile synthesis, multi-layered structure, and high-current carrying capacity. Pertinent to the successful implementation of such applications, however, is the need to develop a thorough understanding of the electrical properties of such materials when subject to high applied electric fields. In this work, we undertake a detailed study of high-field electrical properties of mm-scale, lightly-reduced, rGO papers. Our results reveal that the I–V curves exhibit substantial nonlinearity with associated hysteresis that depends strongly on the applied electric field. The nonlinear behaviour which was interpreted using conventional transport models of Fowler–Nordheim tunneling and space charge limited conduction revealed that while these models provided good qualitative fits to our data, they were quantitatively lacking, thus leaving the issue of high-field transport mechanisms in rGO open for debate. Careful I–V cycling experiments with measurement time-delay introduced between cycles revealed that the observed hysteresis contained recoverable and non-recoverable parts that we identified as arising from charge trapping and Joule heating effects, respectively. Time-dependent measurements showed that these effects were characterized by two distinct time scales. Importantly, the Joule heating was found to cause a permanent conductivity improvement in the rGO via the 'current annealing' effect by effectively eliminating oxygenated groups from the rGO. The analysis of the electrical breakdown in our samples resembled a thermal runaway-like event that resulted in premature damage to the rGO. Finally, we investigated the low-field resistivity in the 80 K–300 K temperature range. The reduced activation energy analysis revealed a robust power law behaviour below 230 K, while deviating from this trend at higher temperatures. For samples that received current annealing treatment, a reduced value for the power law exponent was obtained, confirming the effective lowering of disordered regions.

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