Supplementary Information

Reconstructed Ribbon Edges in Thermally Reduced Graphene Nanoribbons

Muge Acik, ¹ Javier Carretero–González, ² Elizabeth Castillo–Martínez, ² Duncan M. Rogers, ² R. Guzman, ¹ Ray H. Baughman, ² Yves J. Chabal^{1,*}

¹Department of Materials Science and Engineering, University of Texas Dallas, Richardson,

Texas 75080, USA

² The Alan G. MacDiarmid NanoTech Institute, University of Texas at Dallas, Richardson,

Texas 75083, USA



Figure S1. Transmission infrared absorbance spectrum of unzipped GONRs from MWCNTs at room temperature. Vibrational modes are shown for hydroxyls (possible COOH and H₂O contribution) (C-OH, 3000-3600 cm⁻¹), ketones (C=O, ~1750-1850 cm⁻¹), carboxyls (COOH and/or H₂O) (~1600-1750 cm⁻¹), sp²-C (in-plane stretching, ~1500-1600 cm⁻¹), epoxides (C-O-C, ~1280-1330 cm⁻¹ and 800-900 cm⁻¹).



Figure S2. Transmission infrared absorbance spectrum of unzipped GONRs from FWCNTs at room temperature. Vibrational modes are shown for hydroxyls (possible COOH and H₂O contribution) (C-OH, 3000-3600 cm⁻¹), ketones (C=O, ~1750-1850 cm⁻¹), carboxyls (COOH and/or H₂O) (~1600-1750 cm⁻¹), sp²-C (in-plane stretching, ~1500-1600 cm⁻¹), epoxides (C-O-C, ~1280-1330 cm⁻¹ and 800-900 cm⁻¹).



Figure S3. X-ray diffraction (XRD) patterns of GONRs unzipped from MWCNTs showing the evolution of interplanar spacing between graphene layers. The reaction starts with only original CNTs dispersed in H₂SO₄ at room temperature (a), (b) after addition of KMnO₄, and after heating at 55°C for (c) 1 min., (d) 5 min., (e) 15 min., (f) 30 min. and at 70°C for (g) 1 min., and continue till a final GONR formation at 70°C for 10 min. (h). The red line is a guideline for the position of the diffraction pattern corresponding to the interwall distance in the starting CNTs.



Figure S4. Transmission infrared absorbance spectra of MWCNTs before and after unzipping using 850 wt.% of KMnO₄ in H_2SO_4 . (a) MWCNTs at room temperature before unzipping, unzipped MWCNTs (b) as-prepared upon oxidation taken from the batch, (c) after washing with DI water, and (d) after treating thermally at 150°C in Ar flow for 2 hours in a furnace ambient.



Figure S5. Infrared transmission differential spectra of thermally reduced GONRs unzipped from MWCNTs (i) and FWCNTs (ii). Changes of functional groups are shown at temperatures: (a) 60-85°C, (b) 85-100°C, (c) 100-125°C, (d) 125-150°C, (e) 150-175°C, (f) 175-200°C, (g) 200-225°C. C-O-C, COOH, C=O, C-O and C=C represent epoxides, carboxyls, carbonyls, ethers and sp²-hybridized C=C, respectively. The negative peaks below baseline (black dotted lines) show the disappearance (loss) and the positive ones above this line represent new formation of oxygen groups with respect to each reference annealing temperature.



Figure S6. Transmission infrared differential spectra of GONRs from unzipped MWCNTs reduced at 225-600°C. Changes of functional groups are shown at temperatures: (a) 225-250°C, (b) 250-275°C, (c) 275-300°C, (d) 300-350°C, (e) 350-500°C, and (f) 500-600°C. C-O-C, C=O, C-O, and C=C represent epoxides, carbonyls, ethers, and sp²-hybridized C=C, respectively. The negative peaks below baseline (black dotted lines) show the disappearance (loss) and the positive

ones above it represent new formation of the functional groups with respect to each annealing temperature.



Figure S7. Transmission infrared differential spectra of GONRs from unzipped FWCNTs reduced at 225-600°C. Changes of functional groups are shown at temperatures: (a) 225-250°C, (b) 250-275°C, (c) 275-300°C, (d) 300-350°C, (e) 350-500°C, and (f) 500-600°C. C-O-C, C=O, C-O, and C=C represent epoxides, carbonyls, ethers, and sp²-hybridized C=C, respectively. The negative peaks below baseline (black dotted lines) show the disappearance (loss) and the positive

ones above it represent new formation of the functional groups with respect to each annealing temperature.



Figure S8. Transmission infrared differential spectra of GONRs from unzipped MWCNTs using 850 wt.% of KMnO₄ and further annealed at high temperatures (600-850°C). Changes of functional groups are given at temperatures: (a) 850-750°C (black), and (b) 750-600°C (brown). A new peak appears at ~800 cm⁻¹ after annealing at 850°C. The vibrational stretching mode of C=C is also shown at ~1595 cm⁻¹. Removal of C-O and C=O/C=C/C-O appears with a negative peak at ~900-1300 cm⁻¹ and ~1300-1630 cm⁻¹, respectively.



Figure S9. Transmission infrared differential spectra of GONRs from unzipped FWCNTs using 850 wt.% of KMnO₄ and further annealed at high temperatures (600-850°C). Changes of functional groups are given at temperatures: (a) 850-750°C (black), and (b) 750-600°C (brown). A new peak appears at ~800 cm⁻¹ after annealing at 850°C. The vibrational stretching mode of C=C is also shown at ~1595 cm⁻¹. Removal of C-O and C=O/C=C/C-O appears with a negative peak at ~900-1300 cm⁻¹ and ~1300-1630 cm⁻¹, respectively.



Figure S10. XRD patterns for MWCNTs before (black) and after unzipping (blue) using 850 wt. % of KMnO₄ in H₂SO₄ and unzipped MWCNTs using additional H₃PO₄ (red).



Figure S11. XRD patterns for FWCNTs before (black) and after unzipping using KMnO₄ in H₂SO₄ with additional H₃PO₄ (blue).



Figure S12. Transmission infrared absorbance spectra of GOFs and unzipped MWCNTs (GONRs) at room temperature. (a) GO flakes (GOFs) and (b) GONRs from unzipped MWCNTs at room temperature after unzipping with a treatment of H_3PO_4 .



Figure S13. Transmission infrared differential spectra of unzipped GONRs (initially oxidized with H₃PO₄) after annealing at high temperatures (600-850°C). Changes of functional groups are given at temperatures: (a) 850-750°C (black), and (b) 750-600°C (red). A new peak appears at ~800 cm⁻¹ after annealing at 850°C. Removal of C-O and C=O/C=C/C-O appears with a negative peak at ~900-1300 cm⁻¹ and ~1300-1630 cm⁻¹, respectively.



Figure S14. Transmission infrared differential spectra of GO Flakes (GOFs) after annealing at high temperatures (600-850°C). Changes of functional groups are given at temperatures: (a) 850-750°C (black), and (b) 750-600°C (red). A new peak appears at ~800 cm⁻¹ after annealing at 850°C. Removal of C-O and C=O/C=C/C-O appears with a negative peak at ~900-1300 cm⁻¹ and ~1300-1630 cm⁻¹, respectively.