## **Supporting Information**

## **Controlled Reduction of Graphene Oxide Using Sulfuric Acid**

Ana Cecilia Reynosa-Martínez <sup>1</sup>, Erika Gómez-Chayres <sup>1</sup>, Rafael Villaurrutia <sup>2</sup>, Eddie López-Honorato <sup>1,3,\*</sup>

- <sup>1</sup> Centro de Investigación y de Estudios Avanzados del IPN, Unidad Saltillo, Av. Industria Metalúrgica 1062, Parque Industrial, Ramos Arizpe 25900, Coahuila, Mexico; cecilia.reynosa@cinvestav.edu.mx (A.C.R.-M.); erika.chayres@cinvestav.mx (E.G.-C.)
- <sup>2</sup> Thermo Fisher Scientific de México, Avenida Morones Prieto 2805 Pte., Monterrey 64710, Nuevo León, Mexico; rafael.arenas@thermofisher.com
- <sup>3</sup> Oak Ridge National Laboratory, Oak Ridge, TN 37831, USA
- \* Correspondence: honoratole@ornl.gov



**Figure S1.** XPS narrow scan of RGO treated with H<sub>2</sub>SO<sub>4</sub> at a) 1.5 M, b) 5M, and c) 12 M and fuming H<sub>2</sub>SO<sub>4</sub> at d) 12 M. The presence of sulfur it can be observed only in the RGO treated with fuming H<sub>2</sub>SO<sub>4</sub> at 12 M, due to the excess of sulfur trioxide (SO<sub>3</sub>) in the acid.



Figure S2. Raman spectroscopy spectra of GO treated with  $H_2SO_4$  in (a) 1.5 M and (b) 5 M concentrations.

Table S1. Position of bands G and D in Raman spectra and Ib/IG ratio for as-produced GO; GO treated with H<sub>2</sub>SO<sub>4</sub> in concentrations of 1.5, 5, and 12 M; GO treated with fuming H<sub>2</sub>SO<sub>4</sub> in concentration of 12 M; and OG irradiated at 74  $\mu$ W/cm<sup>2</sup> for 120 h.

	G	±	D	±	Id/Ig	±
OG	1,591.83	3.28	1,356.04	0.57	1.62	0.17
1.5 M	1,581.77	1.46	1,344.89	0.40	1.66	0.10
5 M	1,584.52	1.97	1,346.39	0.63	1.75	0.32
12 M	1,599.28	2.15	1,357.97	0.61	1.57	0.03
Fuming 12 M	1,599.28	1.70	1,356.04	1.02	1.45	0.13
74 $\mu$ W/cm <sup>2</sup> 120 h	1,593.78	1.93	1,357.30	0.46	1.83	0.11

	D2	±	D3	±	D4	±	С	±
OG	1,615.99	1.25	1,562.75	3.06	1,141.75	4.02	1,769.99	3.83
1.5 M	1,605.24	0.83	1,548.91	1.46	1,127.07	3.59	1,759.56	3.87
5 M	1,608.78	1.60	1,553.09	3.05	1,128.94	3.19	1,752.25	2.61
12 M	1,621.34	1.68	1,566.05	3.31	1,138.30	2.82	1,763.58	2.87
Fuming 12 M	1,621.34	1.83	1,566.05	2.81	1,138.31	9.51	1,763.58	5.02
74 μW/cm² 120 h	1,616.49	1.16	1,558.95	2.19	1,142.90	1.63	1,750.10	2.82

**Table S2.** Position of bands D2, D3, D4, and C in Raman spectra for as-produced GO; GO treated with H<sub>2</sub>SO<sub>4</sub> in concentrations of 1.5, 5, and 12 M; GO treated with fuming H<sub>2</sub>SO<sub>4</sub> in concentration of 12 M; and OG irradiated at 74  $\mu$ W/cm<sup>2</sup> for 120 h.

Table S3. Values for the FWHM of bands G and D in Raman spectra for as-produced GO, GO treated with H<sub>2</sub>SO<sub>4</sub> in concentrations of 1.5, 5, and 12 M; GO treated with fuming H<sub>2</sub>SO<sub>4</sub> in concentration of 12 M; and GO irradiated at 74  $\mu$ W/cm<sup>2</sup> for 120 h.

	G FWHM	±	D FWHM	±
OG	57.92	1.20	128.16	8.58
1.5 M	61.98	1.46	136.14	1.28
5 M	58.00	1.97	130.51	2.41
12 M	54.12	2.15	135.16	2.05
Fuming 12 M	54.07	1.70	135.16	5.29
74 µW/cm² 120 h	61.88	1.93	148.91	4.78

**Step 1:** Protonation of the hydroxyl group.



Step 2: Loss of a water molecule and generation of a carbocation.



**Step 3:** Formation of a double bond between the carbocation and the neighboring carbon.

![](_page_3_Figure_5.jpeg)

Figure S3. Reduction mechanism of GO by the loss of -OH functional group by the effect of H2SO4.

![](_page_4_Figure_0.jpeg)

![](_page_4_Figure_1.jpeg)

Step 2: Nucleophilic attack of the water towards one of the carbons.

![](_page_4_Figure_3.jpeg)

Step 3: Deprotonation and formation of hydroxyl groups.

![](_page_4_Figure_5.jpeg)

**Figure S4.** Reduction mechanism of GO by the opening of C-O-C functional group (de-epoxidation) by the effect of H<sub>2</sub>SO<sub>4</sub>.