

# Novel Graphene-Based Materials as a Tool for Improving Long-Term Storage of Cultural Heritage

George Gorgolis <sup>1,2</sup>, Steffen Ziemann <sup>3</sup>, Maria Kotsidi <sup>1</sup>, George Paterakis <sup>1,2</sup>, Nikos Koutroumanis <sup>1</sup>, Christos Tsakonas <sup>1</sup>, Manfred Anders <sup>3</sup> and Costas Galiotis <sup>1,2,\*</sup>

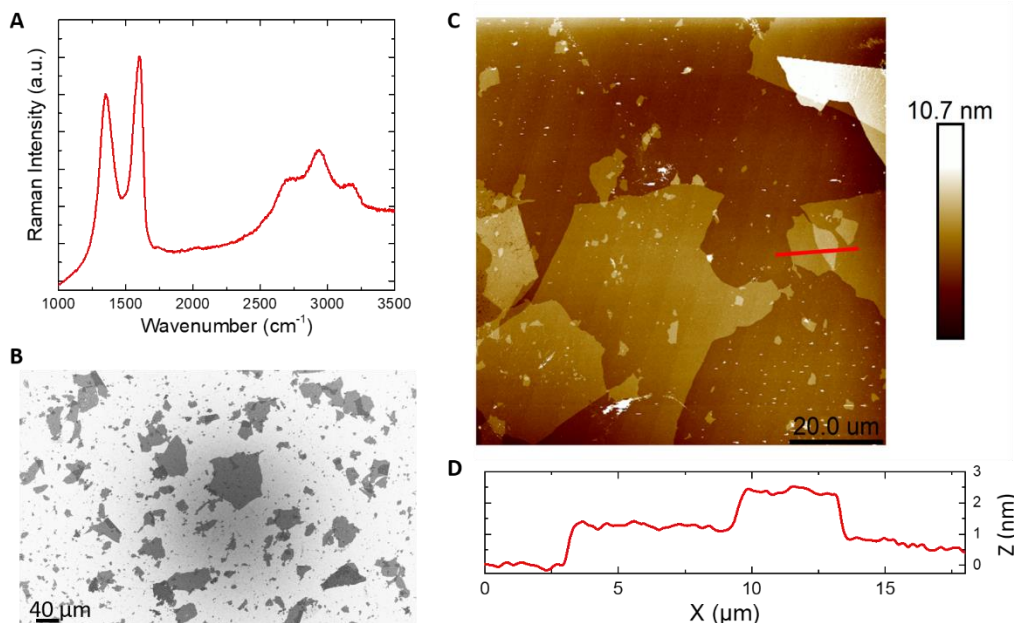
<sup>1</sup> Institute of Chemical Engineering Sciences, Foundation of Research and Technology-Hellas (FORTH/ICE-HT), Stadiou Street, Platani, 26504 Patras, Greece; ggorgolis@iceht.forth.gr (G.G.); kotsidimaria@gmail.com (M.K.); gpaterakis@iceht.forth.gr (G.P.); nickkoutrou@iceht.forth.gr (N.K.); c.tsakonas@iceht.forth.gr (C.T.)

<sup>2</sup> Department of Chemical Engineering, University of Patras, 26504 Patras, Greece

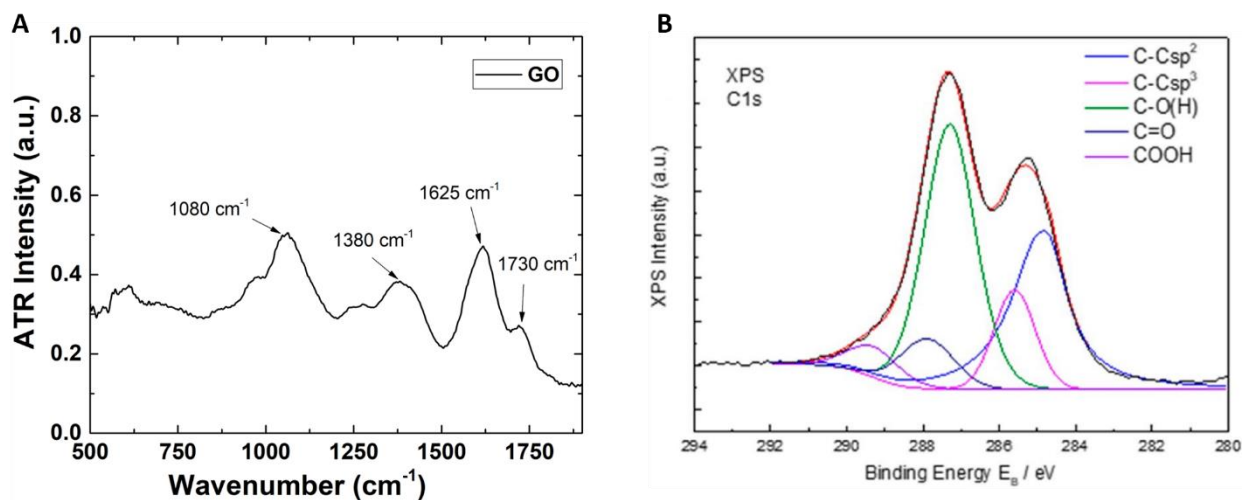
<sup>3</sup> Department of Chemical Engineering, Zentrum für Bucherhaltung GmbH (ZFB), Bücherstraße 1, 04347 Leipzig, Germany; ziemann@zfb.com (S.Z.); anders@zfb.com (M.A.)

\* Correspondence: c.galiotis@iceht.forth.gr or galiotis@chemeng.upatras.gr

The Raman spectrum of GO (Figure S1A) comprises several frequency bands, each one assigned to a certain structural configuration[25]. The main Raman peak is the so-called G peak, located at  $1580\text{ cm}^{-1}$  and is related to the  $E_{2g}$  mode and the in-plane stretching of C=C bonds. The most prominent “disorder” Raman peak is the so-called D peak, appearing at about  $1360\text{ cm}^{-1}$ , is related to the hexagonal ring breathing mode, the intensity of which is associated with the presence of either structural defects or the crystal edges of the lattice [28]. The 2D peak at  $2680\text{ cm}^{-1}$  is the second-order overtone of the D peak, which requires two-phonon scattering for activation, while the other two peaks at  $\sim 2950\text{ cm}^{-1}$  and  $\sim 3100\text{ cm}^{-1}$  correspond to the overtone of the D and G peaks and the G peak, respectively [29]. The two main Raman peaks of GO (G and D) are very sensitive to small changes in the  $sp^3/sp^2$  ratio. Additionally, the intensity ratio of these two peaks,  $I_D/I_G$ , is correlated with the changes of the  $sp^3/sp^2$  ratio and can be used for the characterization of GO. By analyzing a sufficient number of the Raman spectra, the intensity ratio  $I_D/I_G$  for the GO was found to be  $0.97 \pm 0.01$ .



**Figure S1.** (A) Raman spectrum of produced GO, (B) SEM photo for GO sheets, (C) Atomic Force Microscopy (AFM) images of single-layer GO flakes, and (D) height profile of the GO flakes with a thickness of 1.25 nm.



**Figure S2.** Spectroscopic characterization of GO by (A) ATR spectrum [4], and (B) XPS [5].

## References

25. Sygellou, L.; Paterakis, G.; Galiotis, C.; Tasis, D. Work Function Tuning of Reduced Graphene Oxide Thin Films. *J. Phys. Chem. C* **2016**, *120*, 281–290. <https://doi.org/10.1021/acs.jpcc.5b09234>.
28. Ferrari, A.C.; Meyer, J.C.; Scardaci, V.; Casiraghi, C.; Lazzeri, M.; Mauri, F.; Piscanec, S.; Jiang, D.; Novoselov, K.S.; Roth, S.; et al. Raman spectrum of graphene and graphene layers. *Phys. Rev. Lett.* **2006**, *97*, 187401. <https://doi.org/10.1103/PhysRevLett.97.187401>.
29. Das, A.; Chakraborty, B.; Sood, A.K. Raman spectroscopy of graphene on different substrates and influence of defects. *Bull. Mater. Sci.* **2008**, *31*, 579–584. <https://doi.org/10.1007/s12034-008-0090-5>.
20. Schröder, U.A.; Gränäs, E.; Gerber, T.; Arman, M.A.; Martínez-Galera, A.J.; Schulte, K.; Andersen, J.N.; Knudsen, J.; Michely, T. Etching of graphene on Ir(111) with molecular oxygen. *Carbon N. Y.* **2016**, *96*, 320–331. <https://doi.org/10.1016/j.carbon.2015.09.063>.

12. Paterakis, G.; Vaughan, E.; Gawade, D.R.; Murray, R.; Gorgolis, G.; Matsalis, S.; Anagnostopoulos, G.; Buckley, J.L.; O'Flynn, B.; Quinn, A.J.; et al. Highly Sensitive and Ultra-Responsive Humidity Sensors Based on Graphene Oxide Active Layers and High Surface Area Laser-Induced Graphene Electrodes. *Nanomaterials* **2022**, *12*, 2684.