

## ON THE MECHANISM OF CONVERSION OF GRAPHITE TO GRAPHENE OXIDE: INSIGHTS FROM COMPUTATIONAL QUANTUM CHEMISTRY

*Ljubisa R. Radovic,<sup>1,2</sup> Camila V. Mora-Vilches<sup>1</sup> and Ignacio Martín-Gullón<sup>3</sup>*

*University of Concepción (Chile), Penn State University (USA) and University of Alicante (Spain)*

Graphite oxide is normally produced from flake graphite or its parent expanded graphite, a macrocrystalline material when separated by flotation and subsequently thermally purified and partially exfoliated. This layered material has a broad particle size distribution and contains many structural defects. In the classical Hummers-Offeman methodology, it is easily intercalated with anhydrous or highly concentrated H<sub>2</sub>SO<sub>4</sub>. Due to low sulfate oxidation potential (0.17 eV vs. 0.96 for nitrate, 1.70 for permanganate and 1.78 for hydrogen peroxide), this yields pristine graphite oxide whose flakes remain essentially intact; the most often cited oxygen surface groups in this intermediate product are quinones, organosulfates and some sp<sup>3</sup> C-O groups. Be that as it may, the crucial issues regarding the final structure of graphene oxide, and the emergence of its two components, are the relative importance of transfer of one vs. two oxygen atoms to the carbon active sites and the resulting fate of quinone vs. peroxy vs. dioxiranyl surface functionalities. By combining the analysis of results from a battery of complementary experimental techniques with insights from density functional theory, remarkably consistent mechanistic patterns in graphite-to-graphene oxide conversion have emerged. Not unexpectedly, they agree with trends that are reasonably well understood on the basis of surface chemical behavior of sp<sup>2</sup>-hybridized carbon materials ranging from coal or activated carbon to carbon black or graphite. The main chemical features of the graphene-based structures that are formed in this process will be discussed.