

Ever-changing nature of graphene oxide

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Graphene oxide (GO)—the oxidized form of graphene—is actively studied in various fields. One major drawback of GO is its instability [1], which leads to the difficulties in product management. Despite their importance, the structural changes that occur in colloidal dispersions of GO has not been well understood. Optical absorption spectral and magnetic property analyses of GO can provide structural information on the electronic state and sp^2/sp^3 ratio of GO. We accelerated the ripening process of GO by elevation of the temperature to elucidate the unstable nature from the structural aspect. This presentation will show the structurally different GO three states exhibiting inherent magnetic and electrical properties. A physicochemical understanding of the ever-changing nature of GO can promote the establishment of GO science

GO was prepared through the improved Hummers' method. GO dispersions were ripened at different temperatures of 298, 308, 333, and 348 K for up to 2 weeks and an ultraviolet–visible (UV-vis) absorption spectrum of GO dispersions was measured. Magnetic and electrical properties of the freeze-dried GO powders were investigated and the freeze-dried GO powders were also characterized by X-ray diffraction and X-ray photoelectron spectroscopy. Transmission electron microscopic (TEM) images of the freeze-dried GO powders were observed after annealing at 2073 K in Ar. Electron energy-loss spectroscopy was conducted on a specific area on the TEM image.

The three GO states, intrinsic, metastable and transient GO states, are identified using a $\pi-\pi^*$ transition peak of UV-Vis absorption spectra [2]. The intrinsic GO exhibits a $\pi-\pi^*$ transition at 230.5 ± 0.5 nm and stable only for 5 days at 298 K. The composition of oxygen functional groups from XPS measurement and magnetic properties changes with ripening. The paramagnetic nature of GO dynamically changes with ripening (Figure 1). The ripening of GO reduces the radicals on the GO sheets. An optimum choice among the three states of GO can accelerate GO research and applications.

[1] Kumar, P. V., Bardhan, N. M., Tongay, S., Wu, J., Belcher, A. M. & Grossman, J. C. *Nature Chem.*, **6**, 151-158 (2014).

[2] H. Otsuka, K. Urita, N. Honma, T. Kimuro, Y. Amako, R. Kukobat, T. J. Bandosz, J. Ukai, I. Moriguchi, K. Kaneko, *Nat. Commun.* **15**, 1708, (2024).

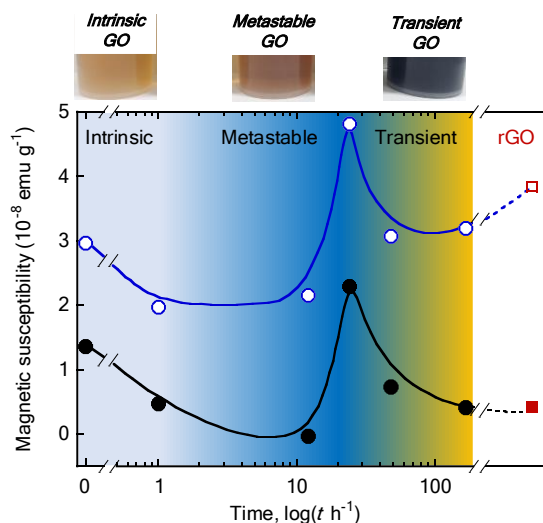


Figure 1. Magnetic property changes with ripening and corresponding GO color.