
I. Rucandio¹

M. B. Gómez-Mancebo¹, R. Fernández-Martínez¹, J.J. Gandía¹, J. Cárabe¹, S. Fernández¹, A. Molinero¹, A. Boscá², J. Pedrós², J. Martínez², F. Calle², F. Leardini³, J.F. Fernández³, A.J. Quejido¹.

¹CIEMAT, Avda. Complutense 40, Madrid, Spain

²ISOM (ETSIT-UPM), Ciudad Universitaria s/n, Madrid, Spain

³Dpto. de Física de Materiales M-04, Facultad de Ciencias, Universidad Autónoma de Madrid, Cantoblanco 28049, Madrid, (Spain)

mariabelen.gomez@ciemat.es

Optimizing the methodology for the production of reduced graphene oxides (rGO) by laser irradiation, chemical, thermal, and combined routes

Graphene has multiple potential applications [1], due to its unique structure, with extraordinary electrical, optical, mechanical or thermal properties. Some of these applications include energy generation and storage, electronic devices, nanocomposites, supercapacitors and so on [1].

Several approaches have been developed for producing graphene [2] and graphene based materials. These graphene based materials have paid a lot of attention because of their versatility, lightweight, abundance and their large surface-to-volume ratio in some cases. One of the most promising graphene-like materials in terms of high yield, low cost and easily up-scalable is the reduced graphene oxide (rGO) produced by chemical routes. The strategy for rGO chemical production is based on the oxidation of graphite to graphite oxide (GO) followed by exfoliation to obtain graphene oxide. Subsequent reduction processes afford a variety of rGOs with tunable Carbon-to-Oxygen (C/O) ratios depending on the reducing agent and reaction conditions although never results in complete removal of oxygen-containing groups [3]. Since the presence of residual functional groups and defects can alter the structure and properties of reduced graphene oxide (rGO), the application of feasible reduction strategies to obtain high quality rGO with low number of defects is crucial [4]. In this work, some routes have been proposed and optimized for reduction of GO for comparison and optimization purposes. These approaches include laser irradiation [5], ascorbic acid addition [6], thermal expansion [7,8] and combined chemical and solvothermal reduction [9]. rGOs from each approach were characterized chemical, structural and morphologically in order to obtain precise information to apply these rGOs to the appropriate application.

GO was prepared following the Hummer's method [10] with slight modifications. Laser irradiation was made using an infrared laser that reduces the graphene oxide [5], converting the initial graphene oxide compact layer into a three dimensional open network of exfoliated graphene flakes. Synthesis of rGO by ascorbic acid as green reducing agent (rGO-AA) was carried out by dispersing of GO in ultrapure water and reduction at room temperature. Thermal reduction and exfoliation was simultaneously performed inserting graphite oxide in an electric furnace. For the solvothermal assisted reduction, a GO aqueous solution was treated with L-ascorbic acid, transferred to PTFE-lined autoclaves and heated at 120 °C for 24h.

The obtained rGO samples were characterized by XRD, SEM, BET and other techniques. These characterizations were performed in order to obtain adequate information to select the best application: high conductivity for electronical applications and high surface area to volume ratio for pollutant removal or energy storage (hydrogen, batteries and supercapacitors). One of the promising applications is the hydrogen storage, in which high surface area is highly recommended. Wherefore, BET analysis provides surface areas information, important parameter in the sorption capacities of these rGO samples (table 1). Laser irradiation treatment has provided the high surface area.

Table 1. BET analysis of rGOs samples reduced by different methods.

| Sample | Specific surface area (m ² g ⁻¹) |
|-------------|---|
| rGO-AA | 37 |
| rGO-AA-T | 27 |
| rGO-thermal | 338 |
| rGO-Laser | 742 |

Acknowledgements

Supported by MINECO projects DIGRAFEN (ENE2017-88065-C2-1-R; ENE2017-88065-C2-2-R) and ECOSGAS (ENE2016-75811-R).

References

- [1] W Choi, I Lahiri, R Seelaboyina, YS Kang. *Crit. Rev. Solid State.* 35(1) (2010) 52-71.
- [2] PT Yin, S Shah, M Chhowalla, KB Lee. *Chem. Rev.* 115(7) (2015) 2483-2531.
- [3] S. Pei, HM Cheng, *Carbon.* 50(9) (2012) 3210-3228.
- [4] S. Thakur, N Karak, *Carbon.* 94 (2015) 224-242.
- [5] A. Ladrón de Guevara, A. Boscá, J. Pedrós, E. Climent-Pascual, A. de Andrés, F. Calle, J. Martínez. *Appl. Surf. Sci.* 467 (2019) 691-697.
- [6] J Zhang, H Yang, G Shen, P Cheng, J Zhang, S Guo. *Chem. Commun.* 46 (2010) 1112-1114
- [7] ST. Nguyen, HT Nguyen, A Rinaldi, NPV Nguyen, Z Fan, HM Duong. *Colloids Surf A Physicochem Eng. Asp.*, 414, (2012) 352-358.
- [8] AV Dolbin, MV Khlistyuck, VB Esel'son, VG Gavrilko, NA Vinnikov, RM Basnukaeva, I Maluenda, WK Maser, AM Benito. *Appl. Surf. Sci.* 361 (2016) 213-220.
- [9] DK Dang, EJ Kim, *Nanoscale Res. Lett.* 10(6) (2015) 1-8.
- [10] WS Hummers, RE Offeman. *J. Am. Chem. Soc.* 80 (1958) 1339-1339.