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Nanosecond Laser-Assisted Nitrogen Doping of Graphene Oxide Dispersions

We have studied different approaches for the synthesis of N-containing reduced graphene oxide (RGO). Thermal treatments of graphene oxide (GO), prepared by a modified Hummer's method, in presence of ammonia gas have been proposed as a simple, efficient and reproducible method for the synthesis of N-doped RGO.[1] These samples are markedly more stable against thermal oxidation in air than their non-doped counterparts, which opens up new possibilities for tailoring the properties of graphene and related systems.[2]

Moreover, we have explored the preparation of N-doped RGO in bulk form by laser irradiation of GO, further expanding the protocols available for the synthesis of N-doped graphene-related materials. The GO sample, dispersed in an aqueous solution of ammonia, was irradiated with a pulsed Nd:YAG laser with emission wavelengths in the infrared (IR) 1064 nm, visible (Vis) 532 nm, and ultraviolet (UV) 266 nm spectral regions (Figure 1).

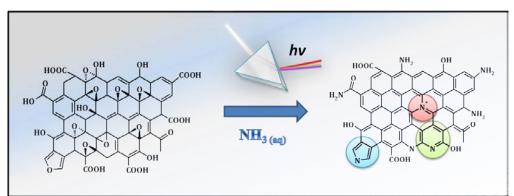


Figure 1. Schematic representation of the synthesis of bulk N-doped reduced graphene oxide samples by laser-assisted synthesis.

We have investigated the role of both the laser wavelength and fluence on the resulting material by means of TEM, XPS, Raman and UV/Vis spectroscopies. Regardless of the laser energy employed, the resulting N-doped material presents a higher fraction of pyrrolic nitrogen compared to nitrogen atoms in pyridinic and graphitic coordination, reaching relatively high N-pyrrolic contents (71.4 %). Noticeably, whereas increasing the laser fluence of UV and Vis wavelengths results in an increase in the total amount of nitrogen, up to 4.9 at. % (UV wavelength at 60 mJ cm⁻² fluence), the opposite trend is observed when the GO is irradiated in ammonia solution through IR processing. The obtained level of doping is within the range achieved by most doping strategies. However, the proposed laser-based methodology allows the bulk synthesis of N-doped RGO in a simple, fast (nanoseconds), and cost efficient manner.[3]

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