Multi-Stimuli Responsive Graphene Oxide For Sensing Application

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Graphene, as a unique 2D nanocarbon material, has kindled great interest due to its extraordinary electronic, thermal and mechanical properties, with intensive promising applications in nanoelectronic devices, sensors, and nanocomposites. Graphene oxide (GO), with the same one-atom thickness but also many oxygenated defects, offers a rather suitable platform for covalent and non-covalent functionalization, owing to the great surface activity and good solution processability [1]. The present work tackles the functionalization of GO sheets to fabricate multifunctional nanoplatforms whose assembly states can be controlled in response to different stimuli for in-vitro diagnostic application.

Specifically, a versatile and smart GO-based substrate - responsive to pH (chemical stimulus) and temperature (physical stimulus) was prepared by covalent grafting respectively of acrylates and acrylamides for interaction with biomolecules [2]. The cooperative effects due to the manifold functionalization were scrutinized with a multitechnique approach of spectroscopic (Raman, X-ray photoelectron spectroscopy, UV-visible, fluorescence) and microscopic (confocal and atomic force microscopy, scanning electron microscopy) methods, both in solution and at the solid surface. In addition, First Principle and Molecular Dynamics calculations were performed in parallel with the experiments.

Results indicate the effective grafting on the GO surface of PAA and PNIPAM with a significant increase of roughness, from AFM analyses (Figure 1), as well as the appearance of the characteristic chemical moieties, from vibrational spectra (Figure 2).

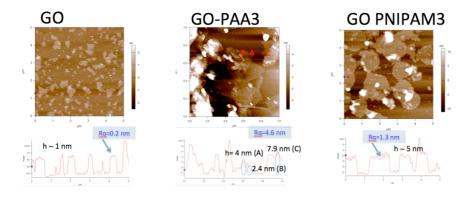


Figure 1: AFM images in AC mode in air of GO sheets and their acrylate (GO-PAA3) and acrylamide (GO-PNIPAM3) derivatives.

Both experiments and theoretical calculations were performed to characterize the response of the acrylamide- and acrylate-GO derivatives (Figure 3).

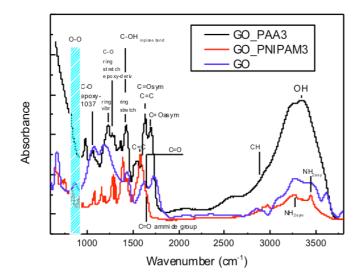


Figure 2: FT-IR spectra of GO sheets and their acrylate (GO-PAA3) and acrylamide (GO-PNIPAM3) derivatives.

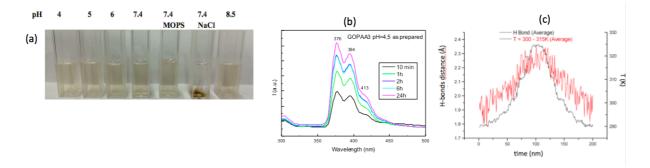


Figure 3: GO aggregation response as function of ionic strength (a), pH (b, GO-PAA3) and temperature (c, GO-PNIPAM3).

The prepared nanoplatforms were tested with a model biomolecule, namely fluoresceinlabeled human serum albumin to assess their optical responsiveness in terms of sensitivity and reproducibility at different ionic strength, pH and temperature values.

In summary, pH- and T-responsive GO nanomaterials were prepared and tested at the hybrid biointerfaces with albumin. Results are very promising for the understanding and control of the processes, both kinetics and thermodynamics aspects, at the graphene-(bio)molecule interface as well as for the (bio)molecule-target interaction in solution.

References

- [1] K.P. Loh et al., *Nature Chemistry* 2 (2010), 1015.
- [2] L. Ren et al., Nanotechnology 21 (2010) 33570
- [3] This research was supported by University of Catania (FIR grant).