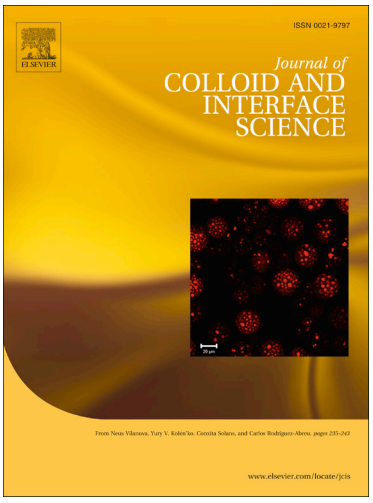

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Surface tailoring of polyacrylate-grafted graphene oxide for controlled interactions at the biointerface

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Abstract

The actual surface termination and lateral size of a nanomaterial is crucial in its interaction with biomolecules at the aqueous interface. Graphene oxide (GO) nanosheets have been demonstrated as promising nanoplatform for both diagnostic and therapeutic applications. To this respect, 'smart' GO nanocarriers have been obtained by the surface functionalisation with polymers sensitive, e.g., to pH, as the polyacrylate (PAA) case. In this work, hybrid GO/PAA samples prepared respectively at low ($GOPAA_{thin}$) or high ($GOPAA_{thick}$) monomer grafting ratio, were scrutinised both theoretically, by molecular dynamic calculations, and experimentally by a multitechnique approach, including spectroscopic (UV-visible, fluorescence, Raman, Attenuated-total reflectance-Fourier transformed infrared and X-ray photoelectron spectroscopies), spectrometric (Time-of-flight secondary ion and electrospray ionisation mass spectrometries) and microscopic (atomic force and confocal microscopies) methods. The actual surface termination, evaluated in terms of the relative ratio between polar and dispersive groups at the surface of the GO/polymer systems, was found to correlate with the average orientation of hydrophilic/hydrophobic domains of albumin, used as model protein. Moreover, the comparison among GO, GO-PAA_{thin} and GO-PAA_{thick} in the optical response at the interface with aqueous solutions, both at acid and at physiological pH, showed that the hybrid GO-polymer platform could be suitable not only to exploit a pH-triggered drug release but also for a modulation of the GO intrinsic emission properties. Energy transfer experiments on the GO/polymer oxide/fluorescein-labelled albumin/doxorubicin assembly showed significant differences for GO and GO-PAA samples, thus demonstrating the occurrence of different electronic processes at the hybrid nano-bio-

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