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Engineering graphene oxide surface chemistry and investigating its effect on the physicochemical and biological properties

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ABSTRACT

The interrelation of physical and chemical properties of GO makes it a very interesting system for several practical applications. It has been widely illustrated that the oxygen content and functionality of GO are intimately related to its physicochemical properties. A lot of focus therefore has been given on tuning the degree of oxidation of GO either by modification of the oxidation conditions of graphite or extent of reduction of as-synthesized graphene oxide. In the current study, systematic tuning of oxidation degree of GO has been explored and a two-step methodology for engineering GO surface chemistry was successfully developed. The investigation of the effects of oxidation degree of GO on its physicochemical properties such as nuclear relaxivity and magnetic resonance imagining (MRI) contrast enhancement and biological properties such as cytotoxicity, drug release kinetics and biomolecule conjugation efficacy was undertaken.
Defect density in the graphite lattice increases with ball-milling time and these induced defects increase the reactivity of the ballmilled graphite. Guided by this, we designed a methodology involving mechanical milling of high purity graphite for different times followed by oxidation of the ballmilled graphite, for synergistically tuning the total oxygen content, relative abundance of different oxygen moieties and size of the resultant GO sheets. The work showcases extensive characterization of the various properties of GO and their interrelation. It also demonstrates how the nature of defects of graphitic precursor can affect the morphology and oxygen content of GO. GO has been used as a nanoplatform for inorganic nanoparticles for designing MRI contrast agents, however the effects of GO on relaxation property of water remains scarcely explored. In this work, the relaxation behavior of GO was probed using nuclear magnetic resonance (NMR) and MRI techniques. Interestingly, the GO samples produced form graphite ballmilled for different times exhibited different relaxivities and different MRI contrast enhancement. The various physicochemical properties responsible for relaxation behavior of GO was investigated and a strong combinatorial effect of manganese paramagnetic ions and oxidation degree of GO on the corresponding relaxivities was noticed. Further, the toxicological effect of the GO series was studied. All GO were loaded with doxorubicin as model drug and the drug release kinetics were observed at different pH. The different GO samples were covalently conjugated with Hyaluronic acid (HA), the model biomolecule taken for the study, and the conjugation efficacy of GO was inspected. Studying these properties can facilitate development of GO based multifunctional agents for cancer cell imaging, targeting and therapy.