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Molecular Dynamics Simulation of the Effect of Surface Charge Density and Oxidation Degree on the Colloidal Stability of Graphene Oxide

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Graphene oxide (GO) nanoparticles have a high adsorption capacity of heavy metal ions (e.g. Cd, Pb, Cu and As) and have been applied in the remediation of contaminated soils. However, GO nanoparticles are also highly water soluble and themselves toxic, such that it is important to understand their fate and transport in the environment, which is governed by their aggregation and colloidal stability. There are two factors that govern the self-aggregation behavior of GO sheets: (a) their surface charge density, which mainly originates from the deprotonation of carboxylic groups at the edge and promotes electrostatic repulsion of GO sheets; (b) their oxidation degree, which is the oxygen-carbon ratio of the basal surface and promotes stronger inter-layer hydrogen bonding and attraction of GO sheets. However, the collective impacts of those two factors on the aggregation behavior of GO sheets are still poorly understood due to limitations in both experimental characterizations and classical DLVO theory. In particular, the experimentally measured zeta potential, which is a reflection of surface charge density, is sensitive to ion adsorption on the GO surface which complicates the characterization of original charge from GO surface, and classical DLVO theory does not account for the spatial heterogeneity of GO surface charge distribution. To gain insight into the colloidal stability of GO, we carried out molecular dynamics (MD) simulations of GO sheets with different surface charge densities and oxidation degrees in bulk liquid water. Specifically, we examined the potential of mean force of two GO sheets at different distances under different combinations of surface charge density and oxidation state. We also examined the interlayer water structure and kinetic properties (e.g. density map, dielectric constant, diffusion coefficient) to gain detailed understanding of water-GO interactions. Our study shows that the surface charge heterogeneity of GO plays a significant role on its colloidal stability. Also, the degree of GO aggregation depends strongly on the ratio of surface charge density and oxidation degree.