**Understanding the pH-Dependent Behavior of Graphene Oxide Aqueous Solutions:**

**A Comparative Experimental and Molecular Dynamics Simulation Study**

**Objective**

A comparative experimental and molecular dynamics (MD) simulation study was carried out to uncover the mechanisms behind the aggregation and the surface activity of GO at different pH values.

**Note:** I will only focus on the molecular dynamics simulations that were carried out in this research paper.

**Methodology**

Most simulations were carried out under the NPT ensemble (constant number of atoms, constant pressure of 1.0 bar, and constant temperature of 298.15 K), except for those involving water vacuum interfaces, which were carried out under the NVT ensemble to generate a large vacuum space on top of the water phase in the simulation box. The optimized potentials for liquid simulations-all atoms (OPLS-AA) force field implemented in the GROMACS 4.0 software package was used for all simulations. The water molecules were simulated using the standard SPC/E model. The equations of motion were integrated with a time step of 2 fs using the Verlet (leap-frog) algorithm. The velocity-rescaled Berendsen thermostat was implemented to maintain the system at a constant temperature. The pressure was coupled to an isotropic Berendsen  
barostat. Periodic boundary conditions were applied in all three directions. The trajectories, velocities, and forces corresponding to all of the atoms in the system were saved every 10 000 steps (20 ps). The PMF between two fixed parallel GO sheets was calculated by numerically integrating, using the trapezoidal method, the interaction forces required to separate the two GO sheets at various intersheet separations, d. the free-energy change during adsorption was calculated by pulling the center of mass (COM) of a GO sheet from bulk water to the water vacuum interface.

**Findings**

At low pH, the carboxyl groups are protonated such that the GO sheets become less hydrophilic and form aggregates. MD simulations further suggest that the aggregates exhibit a GO-water-GO sandwich like structure and as a result are stable in water instead of precipitating. However, at high pH, the deprotonated carboxyl groups are very hydrophilic such that individual GO sheets prefer to dissolve in bulk water like a regular salt. The GO aggregates formed at low pH are found to be surface active and do not exhibit characteristic features of surfactant micelles.