Structural, Electronic and Vibrational Properties of Sulfur-Functionalized Graphene Materials.

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The global surge in demand for batteries has highlighted the known limitations of Lithium-Sulfur (Li-S) batteries[1]. Our research focuses on sulfur-functionalized graphene (GS), a newly synthesized functional material to overcome these limitations. By using density functional theory (DFT)[2], we elucidate the structural, electronic and vibrational properties of GS materials. We have studied the stability of GS materials by first maximizing sulfur content on monolayer graphene sheets, then by identifying stable sulfur binding configurations, and finally by analysing in detail the electronic structure and bonding characteristics. We achieved a significant milestone by determining a stable structure with atomic sulfur content of 20% that aligns with our experimental findings both in atomic percentage and in electronic structure. We have also identified specific vibrational spectroscopic signatures of GS by simulating the Raman spectra of these novel materials. This facilitates a direct comparison between theoretical predictions and our experimental characterization results. The thermal stability of GS materials was considered through *ab initio* molecular dynamics (AIMD) calculations. Finally, we investigated the interaction of GS materials with lithium ions and its behaviour under external electric fields, revealing enhanced electrochemical properties. We have compared the results obtained on graphene to other carbon-based nanostructures, such as carbon nanotubes, fullertubes, and bilayer graphene.

References

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[2] Pykal, M.; Jurečka, P.; Karlický, F.; Otyepka, M., Modelling of graphene functionalization. Physical Chemistry Chemical Physics 2016, 18 (9), 6351-6372

Figures

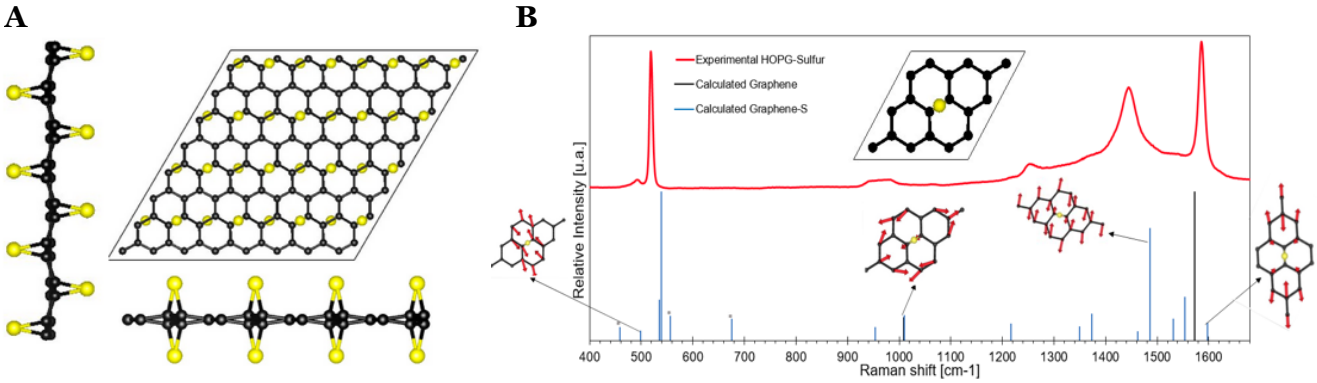

Figure 1: (A) GS (20% atomic S) atomic structure. (B) Simulated (blue) and experimental (red) Raman spectra of Sulfur functionalized graphene.